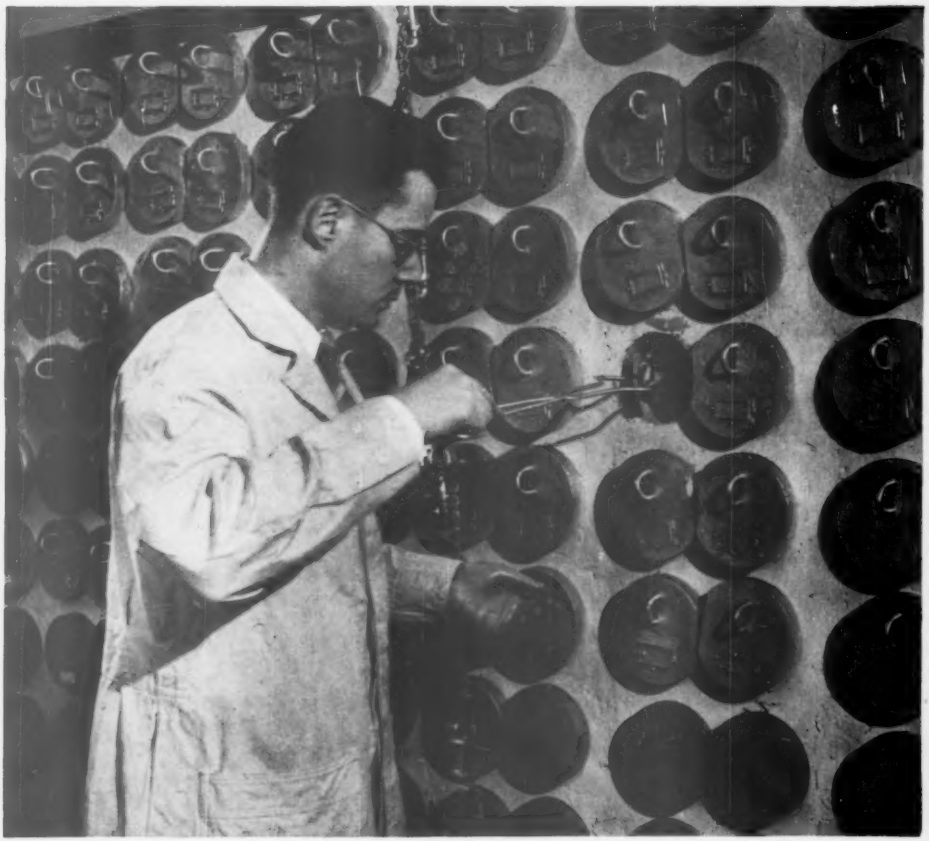
NUCLEONICS



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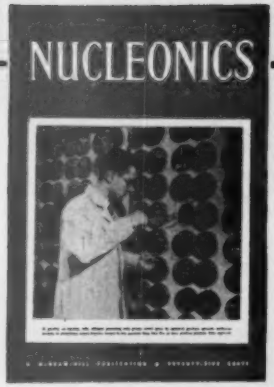

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THE ABC SYMBOL which is printed at the head of this page is, in a very real sense, *your* brand on this magazine. Those letters stand for Audit Bureau of Circulations. The symbol indicates that the magazine is a member and supporter of that Bureau.

To the advertiser who contemplates using the magazine as an advertising medium, this symbol has a well-recognized significance. It tells him that the circulation records and practices of the magazine are wide open to the auditors of the Bureau, who check the publisher's claims and make public the precise terms and conditions under which subscriptions are obtained. And it assures him that the magazine stays in business by virtue of a demonstrated demand from its readers as shown by their paid subscriptions or newsstand purchases.

BUT HERE we are concerned only with the significance of ABC to you as a reader. For when the advertisers, the advertising agencies, and the publishers founded the Bureau nearly forty years ago to help establish honest circulation figures, they unwittingly set up a cooperative institution that has become a major safeguard for the interests of the reading public.

That is because membership in ABC constitutes one of the strongest guarantees that any publication can offer of its primary devotion to the interests of its readers. And by making that guarantee possible, ABC becomes a major safeguard of the freedom of the press, an objective of exceptional importance in these days when the public is flooded with propaganda from so many sources.

THE SUREST MEANS by which to preserve a free press is to keep it directly answerable to the reading public it would serve. It follows, then, that the survival of a truly free press must depend on its acceptance by that public; and that means in turn that the people must have in their hands some adequate means for holding the publishers responsible to them.

No one has yet devised any means to that end more simple, more direct or more practical than the paid subscription or newsstand purchase price. The right to purchase or refrain from purchasing a publication gives to the readers and to no one else the power to pass judgment on whether that publication should continue to serve the reading public.

TO SUPERVISE this vital process, to check and certify the integrity of the publication's circulation methods and claims, requires a strict and continuing audit of each publication's success in meeting this test of its public acceptance. To that essential function the ABC has contributed mightily by the conscientious performance of its mission. And that is why we are able to have a press supported, for the most part, by advertising revenues, but not controlled as to its circulation or content by any influence other than its readers.

When an advertiser consults the ABC statement of a publication to ascertain the amount, the quality and the trend of its circulation, he does so in the legitimate pursuit of his own interest. But at the same time, inevitably, he is helping the ABC to keep the press responsible and responsive to the reading public. For, in effect, he is asking the publication to demonstrate through its circulation figures that it owes its standing to a voluntary demand by its readers.

SO THE Audit Bureau of Circulations, by auditing and certifying paid circulations, has come to perform a vital service to the readers of this magazine and of every other member publication. And in performing that service, it helps to maintain in our country a press that is answerable to the reading public and to it alone. So long as the practices and principles for which ABC stands continue to prevail in American publishing, we shall find in it a sure support for a truly free press, responsible only to the public it serves.

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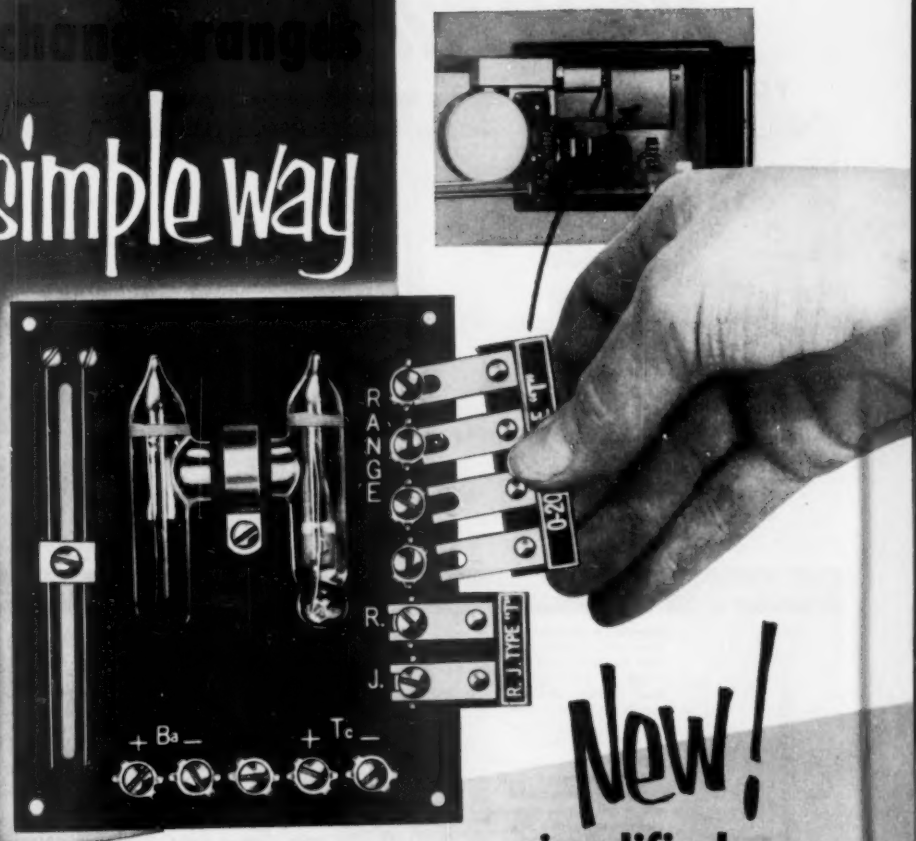

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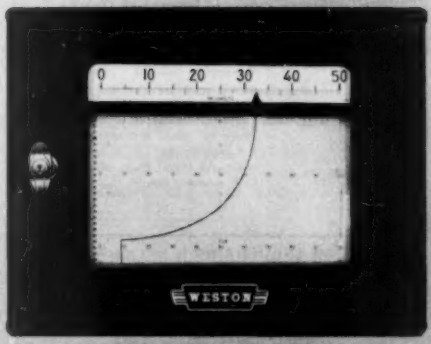

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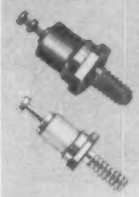

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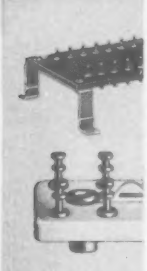

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Ceramic. Silicone impregnated. Type X1986 with 8 lugs staked in two rows. Standoffs riveted and soldered to ground strap for good grounding at R.F. frequencies. 1 1/4" long, 3/8" wide. All metal parts plated. O.A. mounted height: 35/64".

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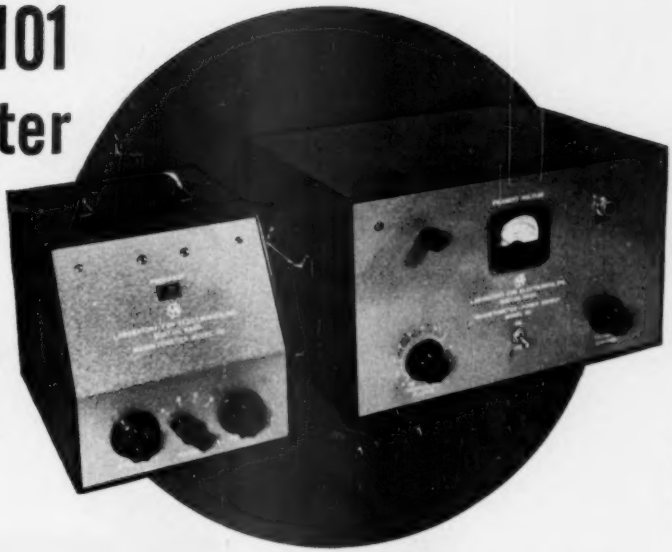

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The Magnetometer translates the problem of the measurement of magnetic field strength to the much more accurately solved one of the measurement of a radio frequency. An oscillatory magnetic field is provided by means of a coil surrounding a sample which permits measurement of proton resonance and the nuclear resonance of lithium (Li^7). The coil is part of an oscillator which is so designed that its level of oscillation drops with an increase in circuit losses, such as may be introduced by nuclear resonance in the sample material. Means are provided to make this resonance easily viewed on an oscilloscope.

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SPECIFICATIONS

Range of Magnetic Field Strength
Proton resonance: 300-8000 gauss
Lithium resonance: 7000-25000 gauss

Frequency to Field Ratio
Protons: 4257.76 ± 0.10 ∿/ gauss
Lithium (Li^7): 1654.61 ± 0.10 ∿/ gauss

Modulation Sweep Width Range
1.6 to 16 gauss

Power Requirements
115 volts, 60 cycles, 30 watts

Frequency Range
Probe 1: 1.18 to 2.84 mc Probe 2: 2.61 to 6.25 mc
Probe 3: 5.6 to 13.5 mc Probe 4: 13 to 34 mc

Area of Sample Material
Probe 1: 0.5 sq. cm. Probe 2: 0.3 sq. cm.
Probes 3 and 4: 0.32 sq. cm.

Size of Probe and Cable
Cable: 18" long Probe: ¾" Diam. ⅝" W.

Size of Units
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APPLICATIONS — Measures temperatures from 3200 to 7000° F. . . . Works with objects as small as $\frac{1}{4}$" in diameter . . . Needs only $\frac{1}{4}$" hole for sighting at target.

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New use of radioactivity helps make better asphalt roads

Operator at Atlantic Refining Co., inserts alpha detecting phosphor by lifting chimney containing 5819 photomultiplier tube. Using different phosphors the G-E Scintillation Counter can also detect beta, gamma, and thermal neutron radiation.

Using a G-E Scintillation Counter to determine beta radiation, The Atlantic Refining Company, Philadelphia, has a new method for measuring the adhesive quality of asphalt which determines the degree to which a road holds together.

This is the test method used: Crushed paving stones are soaked in radioactive calcium chloride solution and dried. The stones are then coated evenly with asphalt, which may contain additives to improve its adhesive characteristics. This mixture is covered with water for a definite period of time.

If the asphalt separates from the stones, the radioactive calcium chloride will dissolve into the water. When the water is evaporated, radioactive residue will remain. The amount of radioactivity is measured with the G-E Scintillation Counter, indicating the resistance of the stone and asphalt to deterioration by water.

Here is a typical example of how industry can make use of the by-products of atomic energy with the help of special instrumentation developed by General Electric.

For more information about this and other G-E Radiation Instruments contact your nearest G-E Apparatus Sales Office, or write for Bulletin GEA-5735 to General Electric Co., Section 687-105, Schenectady, N. Y.

Radiation Detection Instruments

GENERAL ELECTRIC

NUCLEONICS

OCTOBER, 1952

Evaluation of Teletherapy Sources

HIGH-INTENSITY natural radiation sources have been used for many years for the treatment of tumors. Since the advent of nuclear reactors, a variety of isotopes can be produced which may be better for this purpose.

However, because these newer sources are not yet readily available, we do not have the data needed to decide which source is "best." To get some answers to the questions which exist, twenty-five medical schools have formed a Teletherapy Evaluation Board which will study, in cooperation with the Oak Ridge Institute of Nuclear Studies, the various sources as they become available.

THE PRIMARY JOB of the radiophysicist is to build a maximum and uniform dosage in an irregular tumor mass with a minimum dosage elsewhere. Any three-dimensional pattern of dosage can be developed with a beam of radiation from an external source. The machinery for such a source is called a teletherapy unit.

A uniform dosage in any three-dimensional pattern can be developed if there is complete freedom of rotation of the unit, the body, or both. But tumors are often in nonrigid suspension, the body can be maintained only partially rigid for short periods of time, and its limits of rotation are narrow. Therefore, most of the rotating must be done by the unit, placing a premium on its smallness.

The energy from the source should be high to get maximum penetration of tissue and should be low to get minimum penetration through the source shield. Since tissue has a low atomic number Z and the shield has a high Z, the divergent effects of energy and Z on photoelectric absorption and Compton scatter helps resolve the problem.

At any Z, high-energy photons interact largely by the Compton process. Below million-volt energies the photoelectric process increases absorption in the higher-Z shielding materials. The ideal energy from these physical considerations would appear to be below 1.0 Mev but above 0.2 Mev.

Other requirements are: high gamma dose in small source volume, long useful life, and availability. At the present time practical methods of production are by the thermal-neutron reaction and fission-product separation. Depending upon the definition of "high," "small," "long" and "practical," about 6 to 10 radioisotopes are better than radium. Only three appear "practical" for large units: $Eu^{152-154}$, Co^{60}, and Cs^{137}.

About 83% of the gamma energy from $Eu^{152-154}$ is about 1-Mev radiation. The remainder is a mixture of low energies averaging 0.28 Mev and is easily filtered out of the beam. (This data is extrapolated from a narrow-beam absorption in lead and may be inaccurate.) This is essentially the same quality of radiation as that from Co^{60} at 1.2 Mev. $Eu^{152-154}$ will have a specific activity from 10 to 30 times higher than Co^{60} depending upon the conditions of source design and irradiation. The "cold" material is available in both cases and the cost difference is insignificant. Therefore it would appear that $Eu^{152-154}$ is a better source than Co^{60}.

Cs^{137} yields gamma radiation at 0.66 Mev. Its linear absorption coefficient in carbon is 0.18 and compares with 0.14 for $Eu^{152-154}$. Therefore the Cs^{137} radiation is absorbed and scattered only slightly differently than $Eu^{152-154}$ radiation in low-Z tissue-like materials. Cs^{137} should be a few per cent less effective than $Eu^{152-154}$ in depth dose. However in high-Z shielding material the photoelectric component becomes effective. The linear absorption coefficient in lead for Cs^{137} is 1.2 and for $Eu^{152-154}$ is 0.6. A teletherapy unit shield with a Cs^{137} source will be about one fourth the mass of a $Eu^{152-154}$ unit. In a rotating unit under automatic control, such a mass difference is almost decisive. Further, the 33-year half-life of Cs^{137} makes the 5.3-year half-life of $Eu^{152-154}$ seem inadequate. It would appear that Cs^{137} is the logical choice for a teletherapy source.

THERE are a sufficient number of "imponderables" and "overlookeds" to demand experimental verification. The final decision rests with the clinician who assesses the results in patients. Medical statistics being what they are, and since the unit of measurement (degree of palliation) is more a matter of opinion than of fact, the results will be in the form of an opinion poll. One qualified observer's opinion is worth little compared to that of a large group of qualified observers.

MARSHALL BRUCER, *Chief, Medical Division*
Oak Ridge Institute of Nuclear Studies

The Atomic Energy Act and the Power Question

The past year has seen considerable industrial interest in nuclear power. One nontechnical stumbling block is the legal barrier posed by the McMahon Act. Here the former deputy general counsel of AEC outlines changes which might be made in three sections of the Act and points out the importance of the law's keeping pace with technological developments

By BENNETT BOSKEY
Volpe and Boskey, Attorneys-at-Law
American Security Building
Washington 5, D. C.

THE ATOMIC ENERGY ACT OF 1946 (McMahon Act) is now entering its seventh year of active service. To all outward appearance the elements of controversy, indeed rancor, that surrounded its enactment have for the most part slipped quietly into history. Instead, each year's progress in the atomic energy program brings fresh evidence of the constructive nature of the statemanship which guided the law through the two Houses of the Seventy-Ninth Congress.

Experience has demonstrated that in many directions the McMahon Act permits a high degree of flexibility and experimentation. Accordingly, amendments actually enacted thus far by the Congress are few and not fundamental in character. In fact, even the proposals for amendment considered in the Congress have been, on the whole, so modest and so limited as to give further evidence of the general acceptance which the Act enjoys.

But statutory law, we all know, has about it a quality of perishability. Conditions seldom remain static. And an important measure of the success of legislative policy is its capacity for assuring that changed conditions are promptly met by necessary changes in the law.

The McMahon Act gives recognition to this when in a spirit of humility its declaration of policy says:

> "The effect of the use of atomic energy for civilian purposes upon the social, economic and political structures of today cannot now be determined. It is a field in which unknown factors are involved. Therefore, any legislation will necessarily be subject to revision from time to time."

Recently we have heard much of developments in the field of reactors which may, if successfully applied, enable us to realize some of the potentialities of nuclear power. We have heard particularly of proposals for "dual-purpose" reactors designed to produce both plutonium which would be paid for by the Government and power which would be sold to the consumers of power—under arrangements which could make it economically feasible for production facilities to be financed, owned and managed by private enterprise, subject to necessary governmental regulation. Studies being made by industrial teams have led to encouraging statements from both the Atomic Energy Commission and other sources concerning the prospects for dual-purpose reactors.

Should technical progress, even before the next session of the Congress has been concluded, justify undertaking one or more such reactors, these would be developments which may put us for the first time at the threshhold of some major change in the McMahon Act.

What kind of change? In what respects might the present law, with all its breadth and flexibility, be unequal to the occasion? What basic problems would be presented under the Act? For answers to these very natural questions it will be necessary to turn briefly to a few provisions of the law.

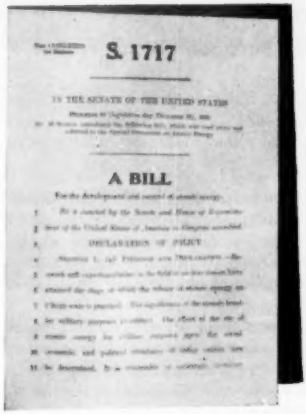
One group of problems arises under Section 4.

Section 4 makes it unlawful for any private person or corporation to own facilities for the production of fissionable material, and it directs that the Atomic Energy Commission shall be the exclusive owner of such facilities. The law excepts from this Government monopoly only those facilities not significant from the standpoint of a weapon—or, in the precise words of the Act, only those facilities which

> "do not, in the opinion of the Commission, have a potential production rate adequate to enable the operator of such facilities to produce within a reasonable time a sufficient quantity of fissionable material to produce an atomic bomb or any other atomic weapon."

The prohibition against private ownership clearly covers the dual-purpose reactor; and it will be noted also that the prohibition is in terms of the capability of the reactor rather than its actual production.

In 1946 the Congress had powerful reasons—five of them were stated in the report of the McMahon Special Committee on Atomic Energy—for incorporating into the legislation so



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A BILL

far-reaching a Government monopoly. It is worth recalling for a moment what those reasons were.

One related to the international situation; hopes then existed for the early achievement of some plan of international control which would be inconsistent with private (or even national) ownership of production facilities in any country. The other reasons related to the nature of fissionable material and the circumstances surrounding its manufacture. For one thing it was believed that private manufacture of material having such enormous destructive potentialities would not be in the public interest. For another, the Congress thought that Government monopoly would minimize the serious hazards to public health and safety which were incidental to the production process. It was felt also that a Government monopoly was more likely to assure continuity of operation. Finally, considerable weight was given to the fact that the production of fissionable material was at that time technologically in its infancy, and that to weaken Government supervision at that stage would be contrary to the principle of prudent stewardship.

It should be borne in mind that at the time the legislation was under consideration the dual-purpose reactor had not emerged as an important possibility, and hence there was no occasion to give specific consideration to the interest which private capital might have in making investment in such a facility. It is true that Section 7(d) of the Atomic Energy Act referred specifically to "byproduct power," and provided that

> "if energy which may be utilized is produced in the production of fissionable material, such energy may be used by the Commission, transferred to other Government agencies, or sold to public or private utilities under contracts providing for reasonable resale prices."

This was essentially an enabling provision which would assure that the Commission was not required to waste any energy produced incidentally as a byproduct in the operation of its installations. It was not directed at dual-purpose reactor proposals as they are now emerging in the light of intervening technical developments. Nor was it intended, as will be seen in a moment from the discussion of Section 7, to constitute a commitment by the Congress to the exclusive use of publicly owned facilities in the production of nuclear power.

With respect to the ownership problem, much has happened since 1946.

First there is the emergence of the dual-purpose reactor as such a distinct possibility.

Second, the domestic position has a somewhat different complexion, resulting from six years of additional experience. During this period the Commission has carried on the production of fissionable materials entirely through a system of contractor operation, rather than by direct operation. A great deal has been learned about subjects such as production techniques and capacities, efficient use of scarce production materials, control and inventorying of special materials, health and safety hazards, labor relations in atomic installations.

And as to international control, the possibilities have altered markedly; if solutions are to be found they must very likely be searched for in directions not contemplated in 1946.

Today we are in a position to make some new assessments. Any consideration of the dual-purpose reactor will entail a fresh inquiry as to whether private ownership of a power-producing reactor capable also of producing fissionable material might not in some circumstances be in furtherance of the public interest. It is true that where the sole object of the activity is to produce fissionable material for which the AEC is the sole customer—such as at the Hanford production reactors —private capital probably would have little or no incentive to invest funds in the facilities, and an attempt to operate on the basis of private ownership would hardly make sense. But with respect to power-producing reactors (whether or not of the dual-purpose type) there would appear to be possibilities of incentives and commercial opportunities comparable to those which have attracted private capital into huge investments in conventional power installations.

The necessity for strict Government control of all fissionable materials remains obvious. But the question is whether this requirement could not be met adequately if a power-producing reactor (capable also of producing fissionable material) were to be privately owned but subjected to a system of carefully developed governmental regulation.

Another suggestion has been for AEC to finance the construction of the reactor and the chemical processing plant and for industry to finance the power plant equipment.

On all such questions the experience of the past six years should contribute much toward an informed and more realistic judgment.

The same developments that make it desirable to reconsider Section 4 bring to life a great variety of problems under Section 7. This is the part of the Act, dealing with utilization of atomic energy, which contains a sort of tentative legislative set of standards for the issuance of licenses by the AEC.

Section 7 requires that

> "whenever in its opinion any industrial, commercial, or other nonmilitary use of fissionable material or atomic energy has been sufficiently developed to be of practical value,"

the AEC shall prepare a report to the President concerning this development and shall also include in its report the Commission's recommendations for necessary or desirable supplemental legislation. It is directed that the President shall then transmit this report to the Congress, together with his own recommendations. The Commission is prohibited from issuing any Section 7 license (authorizing either the utilization of fissionable material or atomic energy, or the manufacture of equipment for this purpose) until this report has been before the Con-

A Commissioner Asks Some Questions about Dual-Purpose Reactors • • •

AEC Commissioner Eugene Zuckert feels that more is involved in bringing industry into the nuclear power picture than merely changing the Atomic Energy Act. National defense policy must be considered with the "highest order of vision and statesmanship." The delineation of responsibilities in a unique pattern of cooperation between business and Government must be made at the outset.

However, if private enterprise is to be attracted, he feels that amendment of the present restrictions on private ownership of reactor facilities and the establishment of equitable patent arrangements are inevitable.

In outlining the problems involved he has indicated that AEC is seeking answers to the questions below. These questions are presented along with answers which the Editors of NUCLEONICS feel are representative of the attitude of a number of the companies interested in nuclear power. Actually the answers are based on the position of the Monsanto Chemical Co.

Q. Can an atomic plant be built to contribute to both civilian and military objectives?

A. *The Monsanto Chemical Company has confirmed the technological feasibility of a combined plutonium and electric power-producing nuclear reactor.*

Q. Will developing civilian atomic power divert more fissionable material than prudent from military uses, that is, from use as bombs and as submarine and aircraft fuel?

A. *We feel we can not only produce more plutonium for our national security program, but perhaps also make it at a lower cost to the Government and the Nation.*

Q. Would the U. S. be justified in buying plutonium (from industrial builders of dual-purpose reactors) for stockpiling beyond our needs, forseeable at the time? How should a price be worked out to assure adequate incentive without entailing an unjustified government subsidy?

A. *The risk in investing in atomic power development must not be much greater than normal business risks if it wants to attract those investment funds which are so scarce. That means the industrial power-plutonium reactor must have a reasonably assured market for its products at fair prices during a period long enough to permit the investor to obtain a return of his funds. And unlike normal commercial operations where there may be hundreds or thousands of customers, the Government is the sole purchaser in the case of plutonium. That creates a problem requiring further study and exploration, for the loss of the reactor's sole plutonium customer could have disastrous results.*

Q. What are the rights of the taxpayers as original stockholders, and what are the rights of industry? How are incentives to be provided for private industry with full recognition of the stake of taxpayers in atomic energy activities they have sponsored?

A. *Some of the problems to be overcome (before industry can fully participate in nuclear power development) lie in the sphere of legislative or legal*

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gress for a period of ninety days. Furthermore, the licensing provisions of Section 7, in their present form, are made subject to the overriding prohibition in Section 4 against private ownership of a facility which produces fissionable material.

It is plain from Section 7 that the Congress wished to reserve to itself full opportunity to consider the subject before the Commission put into effect any system licensing private parties to utilize fissionable material or atomic energy. This certainly was a sensible safeguard, in view of the many issues as to legislative policy that might be raised when the day seemed to be at hand for such peacetime uses of atomic energy.

The suggestion that the issues of legislative policy be reexamined now, prior to any formal determination under Section 7, does not grow out of any expectation that the power industry is about to be revolutionized overnight. It is based on the conviction that vast interests will be affected as nuclear power begins to emerge, that the issues involved are complex in their nature, and that simple ready-made solutions are not to be anticipated.

What kind of statutory criteria are needed to assure that a nuclear power program shall get started in the direction that would be most conducive to the public interest? What measures will be necessary or indeed practicable to afford some reasonable equality of opportunity for entrance into the field? Assuming that the basic materials will continue to be in short supply, what standards should be used for determining the areas and the companies to which they are distributed for use in the production of nuclear power?

A related but somewhat different kind of problem arises in connection with what may develop as a whole new industry—manufacture of equipment for use in the facilities producing nuclear power. How is the store of information accumulated within the Government to be disseminated and made available to those companies which wish to enter into such manufacture? In what ways could effect be given to the direction contained in the present albeit tentative standards established by Section 7, whereby the Commission in consultation with the Attorney General is directed to establish conditions in its licenses which will prevent "the growth of monopoly, restraint of trade, unlawful competition, or other trade position inimical to the entry of new, freely competitive enterprises in the field."

Or to turn to another group of

matters. One of them is determining the scope of the prohibition in the Atomic Energy Act against private ownership of facilities for the production of fissionable material. Adequate machinery can and should be found to police private plants which would produce such material. Our Congress must have felt this possible because in the preamble to the present law it declared the policy of the United States to be aimed at the development and utilization of atomic energy for strengthening free competition in private enterprise, among other things, subject only to the paramount objective of assuring the common defense and security of the nation. No threat to our nation's security would necessarily result from private ownership of facilities to produce plutonium and electric power under security safeguards which would be maintained by the Government.

Q. What about the location of nuclear power plants? Because of the influence of plant location on cost and availability of atomic power, how can the Commission's safety criteria be made less stringent?

A. [*Walker Cisler, president of Detroit Edison, says: "An economic nuclear power reactor should have small exclusion area requirements. The conventional power plant may require a site of about 100 acres, whereas a reactor plant might require, under present limitation, an area 100,000 acres or more."*]

Q. At what stage in the cooperative development of atomic power does the private company begin to acquire rights? How do we insure equitable access to the know-how, developed at Government expense, which must be the background for many patent developments?

A. *Patents and inventions dealing solely with the utilization of atomic energy for military purposes should remain the exclusive property of the Government. However, in order to give maximum encouragement to industrial development of atomic power reactors, there should be a modification in the present policy which forbids the granting of patents for any invention or discovery which is useful solely in the production of fissionable material or in the utilization of fissionable material. For example, should there not be some right to the protection of its inventions granted to a company which has, at its own expense and effort, done further research and development work to apply atomic reactors as direct tools and aids for use in the chemical manufacturing industry?*

Q. What should the Government's responsibility be for health and safety within privately operated plants? What arrangements could be established to assure adequate protection of private atomic plants in the interest of national security?

A. *Our thinking assumes that security regulations required in the national interest and safety regulations for personnel and property would continue to be the primary responsibility of the AEC, as is required by the Atomic Energy Act. At the outset, we realized that paramount security and safety requirements were to remain where Congress has put them and where they now are. Accordingly, we have planned possible future operations with that in mind.*

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issues, how is the competitive position of nuclear power to be established in relation to traditional power rates? To what extent would nuclear power be subsidized through absorption of costs by the Government as part of the price it pays for fissionable material output of a dual-purpose reactor? In what agency or agencies should jurisdiction over nuclear power rates be vested?

This should suffice to suggest some of the issues that must be canvassed, and to suggest also that they are issues which cannot be canvassed adequately without a considerable amount of preliminary exploratory work.

A word should likewise be said concerning the patent provisions in Section 11 of this Act. These exclude from the normal patent system certain areas of atomic energy and provide that in those areas, in lieu of the private monopoly of a patent, the inventor shall have a right to statutory award. The principal ground for the action taken by the Congress in 1946 was that a private patent would be meaningless in an area where the Government was given a monopoly. If, however, the Government's monopoly were to be modified by some change in the law, presumably it would be in order to consider whether the patent provisions warranted change. And in any event it might be desirable to consider whether the dual-purpose reactor, particularly in view of its simultaneous production of plutonium and of power, gives rise to special patent problems that ought to receive separate treatment in the law.

Groundwork should be laid now for a legislative reassessment of all such matters under Sections 4, 7 and 11. Naturally these are problems to which some attention has been given within the AEC and on the outside. But they cannot wait for fuller treatment until all the blueprints of a feasible dual-purpose reactor have been completed and are ready to be acted upon. At that date it would be most unfortunate to discover that, for want of sufficient preparatory work, the country was not in a position to make the necessary decisions. The atomic energy program can ill afford the delays which would occur were the law unable to keep pace with technological developments.

If changes are to be made in the law, they may prove to be changes of considerable magnitude. Their implications should be carefully thought out, not merely within the Government but by people on the outside. This process needs to be accelerated if the job is to be done in time.

Measurement of Natural Radioactivity Background

An installation using radioisotopes must, in its monitoring program, take into account the presence of natural radioactivity. This paper outlines the techniques found most effective for air, water, and ground monitoring

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MANY COMPETENT scientists have devoted a major fraction of their investigations to measuring the radioactivity present in the natural environment. Originally, their object was to obtain information on the source of the active materials, and later, their interest lay largely in geological dating and investigation of modes of formation of these materials.

With the advent of mass production of radioisotopes, the interest in such measurements was extended, since the background radiations from natural sources interfere with the delicate measurements required to detect hazardous, or potentially hazardous, quantities of radioisotopes escaping from an operating plant.

Many people assume that a given type of radiation will be hazardous regardless of the particular isotope involved. Unfortunately, however, the degree of hazard varies depending upon the energy and type of radiation, the half-life of the isotope, and the metabolic fate. Thus, the maximum permissible concentration of plutonium in the air is 2×10^{-12} $\mu c/cm^3$, while natural concentrations of radon exceeding 10^{-10} $\mu c/cm^3$ are not uncommon.

Air Monitoring

Three types of radioisotopes are of concern in air monitoring: (1) aerosols, or airborne particulates, containing radioactive materials; (2) vapors or gases which may be metabolized into the body; and (3) noble gases which are not metabolized, although they may have appreciable solubility.

Aerosols. These may consist of actual small particles of the radioisotope or of active materials adsorbed on the surface of dusts. Sizes of particles and the activity carried by individual particles vary widely, depending upon the source. Maximum permissible limits (in terms of maximum permissible concentrations) of several isotopes for workers in the field, as obtained from the recommendations of the International Commission on Radiological Protection (*1*) —along with the detection limits required by these values for several types of samples—are given in Table 1.

These values are calculated assuming uniform irradiation of the organ in which the radioisotopes are deposited with eventual elimination of the isotopes from the lung either by absorption or by complete removal from the body. The problems attendant with very active individual particles which may lodge at a given location and irradiate a small mass of tissue, or the settling of these materials on vegetation, water, etc., later used for domestic purposes, have not been considered in the tabulation.

Interferences with measurements designed to detect effluent contamination arise chiefly from the presence of radon and thoron daughter products. Radon concentrations vary from 5×10^{-10} $\mu c/cm^3$ (*2*) in mountainous regions to 10^{-12} $\mu c/cm^3$ (*3*) over the ocean. Thoron concentrations have not been investigated as extensively, but appear to have maximum values on the order of 4×10^{-12} $\mu c/cm^3$ (*4*). However, this may be exceeded in rooms with walls containing significant amounts of thorium minerals.

Interference of these gases with aerosol measurements arises from the

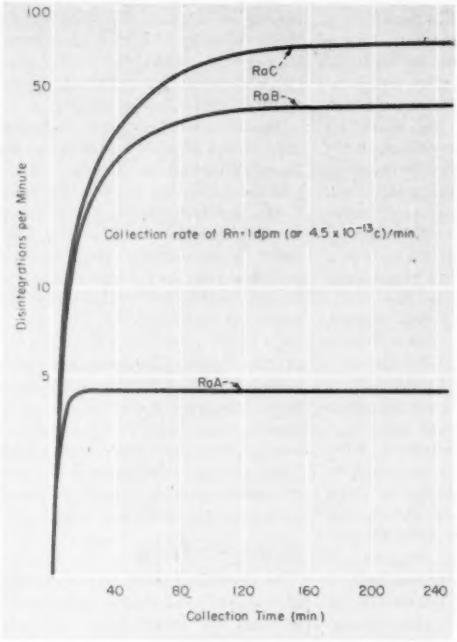

FIG. 1. Build-up of radon daughters on collector

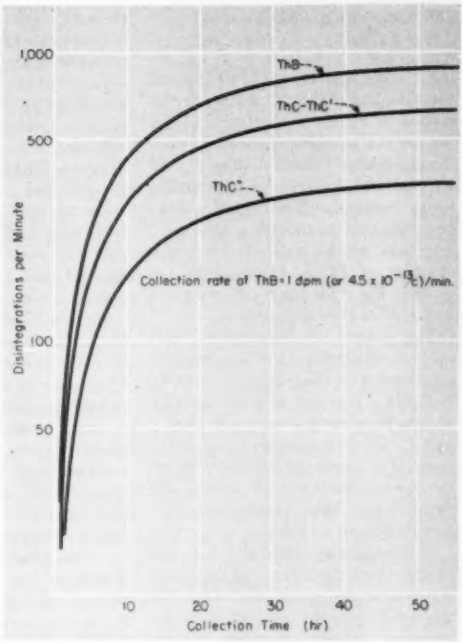

FIG. 2. Build-up of thoron daughters on collector

collection of the daughter products (RaA, RaB, RaC, and RaC') on the filter paper, electrostatic precipitator, or other means used in collection of the dust. Since the daughter products of both chains have comparatively short half-lives (RaB—26.8 min, ThB —10.6 hr), the decay occurring during the collection time may be significant. Correction must also be made for the build-up of daughter products from the parent previously collected. Thus, in the radon chain, the RaC present after a short collection period is due to the RaC that has been collected and the RaC resulting from decay of RaA.

Equations relating the amounts of any daughter to the radon or thoron concentration, assuming uniform concentration in the air and secular equilibrium between the members, are

$$N_A = (FC/\lambda_A{}^2)(1 - e^{-\lambda_A t}) \quad (1)$$

$$N_B = FC[(\lambda_A + \lambda_B)/\lambda_A\lambda_B{}^2 + e^{-\lambda_A t}/\lambda_A(\lambda_A - \lambda_B) - \lambda_A e^{-\lambda_B t}/\lambda_B{}^2(\lambda_A - \lambda_B)] \quad (2)$$

$$N_C = FC[L - Me^{-\lambda_A t} - Qe^{-\lambda_B t} + (M + Q - L)e^{-\lambda_C t}] \quad (3)$$

where L, M, and Q are defined as

Table 1—Maximum Permissible Levels of Aerosols

Isotope	*M. p. c.* ($\mu c/cm^3$)	*Spot sample* *15 ft³/min for 1 hr* (*dpm*)	*Average sample* *10 ft³/min for 1 week* (*dpm*)	*Continuous sample* *2.5 ft³/min* (*dpm/hr*)
Ra^{226}	8×10^{-12}	430	5×10^4	72
Pu^{239}	2×10^{-12}	108	1.3×10^4	19
$Sr^{90}(+Y^{90})$	2×10^{-10}	1.1×10^4	1.3×10^6	1,900
P^{32}	3.6×10^{-8}	2.0×10^6	2.3×10^8	3.3×10^5
Ca^{45}	2.4×10^{-10}	1.3×10^4	1.5×10^6	2,200

Table 2—Collection of Naturally Radioactive Materials from the Air

	Spot sample (*15 ft³/min for 1 hr*)		*Average sample* (*10 ft³/min for 1 week*)	
	α emitters (*dpm*)	*β emitters* (*dpm*)	*α emitters* (*dpm*)	*β emitters* (*dpm*)
Radon				
10^{-10} $\mu c/cm^3$	5,000	8,000	4,500	7,000
10^{-12} $\mu c/cm^3$	50	80	45	70
Thoron				
10^{-12} $\mu c/cm^3$	55	110	630	1,200
10^{-12} $\mu c/cm^3$	6	10	60	120

$$L = \lambda_A\lambda_B + \lambda_B\lambda_C + \lambda_A\lambda_C/\lambda_A\lambda_B\lambda_C^2$$
$$M = \lambda_B/\lambda_A(\lambda_A - \lambda_B)(\lambda_A - \lambda_C)$$
$$Q = \lambda_A/\lambda_B(\lambda_C - \lambda_B)(\lambda_A - \lambda_B)$$

and F is the flow rate of the air through the sampling device; C is the concentration of radon or thoron; N_A, N_B, N_C are the numbers of atoms of each daughter after collection time T; and λ_A, λ_B, and λ_C are the appropriate decay constants. Evaluations of these equations for a collection rate of 1 dpm/min or 4.5×10^{-13} curies/min (product of FC is 1 dpm/min) are given in Fig. 1 for radon collection and in Fig. 2 for thoron collection.

The degree of interference from natural emitters in the air may be estimated from the required sensitivity in Table 1 together with the activity present from natural sources in Figs. 1 and 2. Table 2 presents a brief summary of the amounts of active materials on a collector at the end of the "spot-sample" and "average-sample" periods defined in Table 1.

By comparison with Table 1, it will be noted that the radon contribution in many locations equals or exceeds the amount of activity collected from the maximum permissible concentrations of other isotopes. The thoron daughters are sufficiently high to cause uncertainty in the interpretation of the results. Since it is common practice on all such limits to permit the exposure of the general population to doses of only 1–10% of those quoted,* the problem becomes acute in the environmental monitoring program. This is especially so since the detectable limit should be lower than the maximum permissible by at least one order of magnitude to detect substandard disposal practices as soon as possible.

These active daughters decay, according to conventional decay and build-up relations, to a transient-equilibrium state between RaB and ThB and their daughters. Thus, the effect of radon daughters may be eliminated by allowing the sample to stand for 4–6 hr before measurement. The most satisfactory method of eliminating the thoron daughters, if the active material of interest has a long half-life, is to obtain two measurements at least 10 hr apart after the radon daughters have decayed, and to solve for the fraction of the activity showing no decay in this period.

Measurements of radon and thoron contents in the air may be made by condensing the gases with liquid air and distilling them into an ionization chamber, adsorbing the gases on activated charcoal, or collecting the daughter products on a filter paper or charged wire and applying Eqs. 1–3. Since the concentrations vary widely from hour to hour and from day to day, any evaluation should include a number of samples over a long enough period to indicate seasonal trends.

Metabolized gases and vapors. Metabolized gases or vapors consist of such materials as $C^{14}O_2$ and I^{131}, which may be utilized by the body in metabolism, and thus have maximum permissible concentrations well below those set by external radiation. Since these materials must be monitored by specific analytical techniques involving scrubbers or absorption towers, the effects of natural radon or thoron may be eliminated in the analysis. It should be noted that vapors may in themselves serve as an interfering factor in the aerosol measurement because of condensation or adsorption on dust particles or on the collector.

Noble gases. Since noble gases, such as argon, xenon, and krypton, are not involved in body metabolism beyond their solubilities in body fluids or fats, their maximum permissible levels in air are determined by the external radiation received by the organism. Thus, monitoring techniques consist of total radiation measurements. The limit, as set by the International Commission on Radiological Protection, is 0.3 r per week measured in free air (*1*) or, for environmental measurements in populated regions, some lower values such as 1% or 10% of the 0.3 r per week.

Normal backgrounds for this measurement differ somewhat from one location to another depending upon the radium content of the soil, the elevation, and the natural radon or thoron concentrations. Cosmic rays account for 1–5 ion pairs per sec per cm^3 of air (*5*). Evans has indicated a linear relation between the radium content of soil and atmospheric ionization, with a constant of 5.5 ion pairs per sec per cm^3 of air for a radium concentration in the soil of 10^{-12} gm/gm (*6*). The contribution from natural isotopes in the air, excluding alpha particles which would only register on internal chambers, should be on the order of 0.01–1 ion pair per sec per cm^3. Thus, the total dosage rate due to background should be between 0.1 and 0.7 mrep per day.

Measurements of gross radiation may be made with recording or integrating ionization chambers or, if greater sensitivity is desired, with G-M counters exposed to a large sweep of the atmosphere. A conductivity meter is occasionally used for this purpose, but careful interpretations of the results are needed since the ionization measured by this device may result from heat, electric ares, or other factors. If vapors are present, some method of preventing contamination of the ion chamber or counter is needed. Anomalies in the records of total gamma measurements have been found and attributed to the detonation of atomic bombs several thousand miles from the instrument site (*7*, *8*).

Water Monitoring

Monitoring of water in the vicinity of a plant or laboratory includes the underground water table and any streams or bodies of water into which radioactive effluents may flow. The upper limits of measurement required for soluble materials may be taken from the values recommended for drinking water by the International Commission on Radiological Protection (*1*). Certain of these values are listed in Table 3, along with the estimated response of several instruments to the amount contained in a sample of the water.

These values are calculated on the basis of drinking-water contamination and do not include possibilities of concentration by biological organisms, or adsorption on silts with future transfer to plants or animals. Specific locations may have local rulings which provide more stringent factors to allow for these unknown factors. Again, the maximum permissible levels for public water supplies are considered as 1–10% of the values quoted. In addition, at least one more order of magnitude is desired in sensitivity so that leaks may be corrected before the maximum permissible level is reached.

Natural water supplies contain radium, uranium, and dissolved radon in varying amounts, depending upon the ultimate source of the water and the geological formations encountered in its flow. Even rain water may

*At present, there appears to be an increasing move toward the application of the 10% factor for the exposure of the general populace.

Table 3—Maximum Permissible Concentrations in Drinking Water

Isotope	M. p. c. ($\mu c/cm^3$)	1-cm^3 sample		500-cm^2 sample	
		(dpm)	(cpm*)	(dpm)	(cpm*)
Ra^{226}	4×10^{-8}	0.088		44	
Pu^{239}	1.5×10^{-6}	3.3		1,600	
$Sr^{90}(+Y^{90})$	8×10^{-7}	3.6	0.6†	1,800	300†
Na^{24}	8×10^{-3}	18,000	3,600	9×10^{6}	1.8×10^{6}
P^{32}	2×10^{-4}	440	95	2.2×10^{5}	4.8×10^{4}
Co^{60}	1×10^{-5}	22	1	11,000	440
I^{131}	3×10^{-5}	66	8	33,000	4,000

* For evaporated sample on 1-in. plate, 23% geometry, a 4 mg/cm^2 1¼-in.-diameter mica-window tube, and 200 ppm of solids.
† Includes Y^{90} daughter.

contain measurable amounts of radon and radon daughter products dissolved from the air. Radon concentrations are normally between 10^{-4} and 10^{-8} $\mu c/cm^3$, with the higher values in springs or wells and the minimum values in turbulent streams (*9, 10*). The concentrations in specific springs may be higher than the values quoted by factors of 10–100. Tap water may be somewhat depleted in dissolved radon because of the turbulence and pressures encountered in pumping. Radium concentrations vary widely—some natural springs have concentrations up to 10^{-7} $\mu c/cm^3$ (*11*) and ocean-water concentrations vary from 3×10^{-11} to 2×10^{-10} $\mu c/cm^3$ (*12, 14*). Thorium concentrations are generally less than 10^{-9} gm/cm^3, although significant quantities of $MsTh_1$ may occur (*13*). Most natural waters contain 5×10^{-10} to 3×10^{-8} gm of uranium per cm^3 (*4*).

The degree of interference of these materials with water monitoring is difficult to estimate. The long-lived products are seldom in radioactive equilibrium, and the response of the measuring device depends on the relations between parent and short-lived daughters and, thus, on the time between chemical manipulation of the sample and measurement. As an example, in the gross determination of alpha emitters by counting an evaporated residue, the possibility of the radon daughter products registering on the measuring equipment depends on the initial concentration and the time interval between the start of evaporation and the measurement of the residue. If the sample initially contains 10^{-6} $\mu c/cm^3$ of dissolved radon, a 500-ml sample would then have 1,100 dpm each of RaA, RaB, RaC, and RaC′. Four hours after the radon is separated by the evaporation, there would remain about 8 dpm of beta emitters and 6 dpm of alpha emitters. The alpha emitters, in particular, could give positive indications of contamination, under these circumstances, with some needless concern if it is attributed to the longer-lived, more hazardous isotopes.

Uranium and radium contribute mostly to the alpha-emitter measurements in gross determinations, although uranium is carried by many of the extraction procedures for plutonium. Since uranium has a specific activity of about 1.5 dpm/μg (including U_{II}), about 1.5 beta particles per microgram from the two intermediate daughter products, UX_1 and UX_2, would be expected. In passage through the soils, however, the UX_1 and UX_2 appear to be preferentially absorbed, leaving the resulting uranium with a much lower ratio of alpha-to-beta radiation than is expected. Samples containing radium may show an increase in activity with time when prepared for counting because of radon trapped in the residue or precipitate with subsequent build-up of the daughter activities.

Procedures for the analysis of these materials in water are numerous in the literature. Radon is usually determined by boiling the sample and flushing the gases into an ionization chamber, or counter, with nitrogen, argon or similar gases (*14*). A rapid method, involving conventional radiochemical laboratory equipment, consists of precipitating the radon daughters and measuring the number of beta particles (*15*). Radium is usually determined by sealing a sample and allowing it to stand for 30 days until the radon has built up into equilibrium, and then measuring the amount of radon produced (*14*). Uranium may be determined by microchemical techniques or, more simply, by measurement of the fluorescence of a melt in sodium fluoride under ultraviolet excitation (*16*).

Ground Monitoring

Monitoring of the ground in the region of a laboratory correlates with both air and water monitoring, since the extraneous active materials on soils or plants usually arise from air-borne dusts or vapors or from adsorption from water streams. The three main items involved in ground surveys are soil, vegetation, and, as a final check on the values from the other two, local animal life.

Soil measurements. Since the maximum permissible concentrations of radioactive materials in soils have not been defined and are nebulous, depending upon the use of the soil in the region, it is impossible to state any definite limits of sensitivity for the analysis. In general, the sensitivity should be as good as is compatible with both radiological safety and time spent on the measurement. Thus, for heavily populated regions, considerable effort may be advisable to insure against spread of contamination to water tables or edible plants. In deserted or restricted areas, less effort is needed to insure against the movement of possibly hazardous amounts to other locations.

Soils, unfortunately, contain to some degree nearly all of the more common natural radioelements, along with varying amounts of daughter products—depending upon the source of the sample and the treatment before measurement. Uranium concentrations are quoted up to about 6×10^{-5} gm/gm of soil (*17*) (excluding ore bodies). This would give about 10 alpha and 5 energetic beta dpm/gm of soil, if only the daughter products through U_{II} (U^{234}) are considered.

Thorium occurs in concentrations up to 6×10^{-5} gm/gm of rock (*17*), which could give rise to total disintegration rates of 100 dpm of alpha emitters and 70 dpm of beta emitters if the entire chain is considered in equilibrium in the sample. Average values for thorium would be approximately one fifth of these disintegration rates.

Radium concentrations are normally

COUNTING ROOM at Hanford where environmental samples are measured for β emission

about 10^{-12} gm/gm of soil (*18, 19*), or about 2 dpm/gm of soil. The effects of daughter products may increase this to a total of 10 alpha and 8 beta dpm if it is assumed that the radon has remained trapped in the sample. Both radium and thorium have higher concentration in igneous rocks than in sedimentary rocks.

Potassium contents may be troublesome because of the naturally active K^{40}, which contributes about 30 beta particles per sec per gm of potassium (*20*). Since soils contain 10^{-3} to 3×10^{-2} gm (*21*) of potassium per gram of soil, about 2–50 beta particles per min may be expected.

Measurements of these materials are made by techniques similar to those used for water samples. In most cases, the soil is placed in solution by fusion with sodium carbonate and treatment with hydrofluoric acid.

Radium is determined by sealing for one month and measuring the radon in equilibrium [Evans has described a technique involving melting rocks in an electric furnace and measuring the radon evolved (*14*)]. Thorium may be determined by chemical procedures or by constant flushing of thoron into an ion chamber or counter (*14*). Uranium is measured by the fluorescence of a melt under ultraviolet light, and potassium is determined by a flame photometer (*22*). Many of these materials, when adsorbed on the soil from waste solutions, may be removed by a strong acid, thus eliminating the need for the time-consuming fusion step.

Vegetation. Maximum permissible limits of radioactive materials on vegetation are usually determined by the possibility of ingestion by livestock feeding in the area, although the possibility of human consumption in significant quantities must be borne in mind. Since values for these limits have not been published, the same general statements about required sensitivity that were applied to the soil measurements are valid here.

The main source of interference is the K^{40} associated with the potassium in the plant structure. Naturally occurring plants contain between 1 and 20 mg of potassium per gram of vegetation (*23*). The beta-particle emission would then amount to 2–40 beta particles per min per gm. Indications of a seasonal trend in the potassium content, with maximum values during the growing season, have been noted in the case of desert plants of the Pacific Northwest.

Traces of uranium, thorium, and radium have been reported in plant life, but the amounts are low in comparison to the potassium content.

Biological monitoring. The animal life in the region serves as an admirable check on biological accumulation since the individual organs may be examined. Any significant increase over background, in this case, may be of concern since such a phenomenon, involving livestock or possibly humans, may increase in magnitude if the source is not found and controlled.

Again the chief interfering isotope turns out to be K^{40}. The average potassium content of animals is about 3 mg/gm (*24*) with variations by a factor of 10–20 between types of tissue (*25*). Thus, the average tissue contributes about 5 beta particles per gm of tissue. Fortunately, this value should not vary greatly from locality to locality, so that determinations on a given animal, obtained at a distance from the area of interest, should provide an adequate basis for comparison.

Radium is quoted as 9,000–18,000 dpm in an 80-kg man (*26*). If this is considered as occurring chiefly in bone, a normal value of 0.9–1.8 dpm/gm would be obtained. This value may be subject to some fluctuation from locality to locality because of differences in the natural radium content of the water supplies. The C^{14} content of the body is about 150,000 dpm (*26*) or 1.9 dpm/gm of tissue.

Laboratory Procedures

Unfortunately, the measurement of natural radioisotopes is not the only background problem encountered in monitoring work, since the conditions of the work location will also affect the final result.

Most laboratories doing this type of work cannot, either due to economic or efficiency factors, be located at a distance great enough from the actual operation area to eliminate the possibility of cross-contamination. To assess the degree of such a contribution, it is advisable to process a number of "blank" samples along with the actual samples. These blanks are identical to the samples of interest except that they have not been exposed to radioactive contamination prior to analysis. A source of such samples is sometimes difficult to find, but material obtained at a distance from the operation location, preferably in the same type of country, is acceptable.

Similarly, the efficiency of any chemical procedure should be checked by routine analysis of samples containing known amounts of the isotope of interest. This permits evaluations of losses in the manipulations and insures good consistency from day to day. Comparisons of these two sets of values by

statistical means allows a reasonably accurate estimation of detection levels, and prevents reporting of spurious traces of radioactive materials introduced in the laboratory. These measurements must be performed at the same time as the analyses since conditions may change from time to time. Thus, it is not always possible to estimate the final sensitivity limits of a procedure before a laboratory or operating plant is in production.

Discussion

Measurements of the natural radioisotopes are of value in monitoring work in two cases:

1. Where gross determinations of total particle emission are used to estimate trends of extraneous activity.

2. Where the waste from the laboratory contains the natural isotopes.

In the first case, the contribution from natural emitters may mask the radiations from the isotopes of interest. No estimate of sensitivity levels or the adequacy of a particular measurement can be made until the limits of the natural activities are known. Gross determinations are frequently used to permit rapid examination of a large number of samples, with specific analysis only on those showing deviations from the average.

The second case is considerably more troublesome since it is almost impossible to differentiate between the natural isotope already present and that added by waste disposal, at least without a considerable expenditure of time and money in studying equilibrium conditions between members of the chain both in soil and water samples. If prior knowledge of natural concentrations is not available, an actual waste could be attributed to natural concentrations, and the source of the activity may not be traced until the concentrations have built up to possibly hazardous levels.

A survey of the site before the start of operations permits evaluation of natural isotopes without interference from artificial isotopes. This provides a backlog of data against which the gross analyses proposed for control monitoring may be compared to show sensitivity levels for the contaminating wastes of interest and serves as a basis of comparison for future analyses.

For major locations, this background survey should be only a part of an over-all program which includes geological studies of underground strata and water table; meteorological studies of wind velocities, temperature inversions, etc.; and possibly ecological surveys of the wildlife as a basis for noting changes after operations start.

BOAT USED for collection of water samples and biological specimens at Hanford Works

Particular care must be taken to investigate possible daily, weekly, or monthly trends. One such trend, that of the potassium content of vegetation, has already been noted. Others include the fluctuation of radon and thoron concentrations noted throughout a single day, with possible seasonal changes; and possible changes in uranium or radium in the water tables, particularly when close to a large stream. These changes are undoubtedly dependent to some extent on the particular climatic, hydrographic, and geologic features of the location.

The amount of time devoted to this work, of course, depends upon the size and complexity of the operations. For minor laboratories, background studies are of little value since only one or two isotopes are involved. Any monitoring effort in this case would probably involve specific analyses which exclude natural activities. For major installations, entire groups would be devoted to studies concerning the effect of natural emitters, meteorological conditions, and geological structures. In the latter case, interferences from natural isotopes must be evaluated in order that proper interpretations of the gross measurements may be made.

* * *

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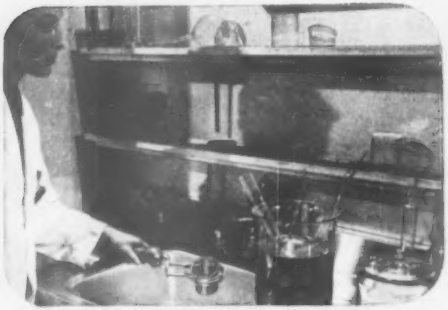
Aid in teaching radioisotope techniques is now available in the form of training films which AEC has produced in collaboration with the Armed Forces Institute of Pathology. Entitled "The Radioisotope," the 13-film series is intended for technicians, doctors, engineers, and scientists.

First completed films show what the series offers in details (a reproduction of Hevesy's lab and plant-metabolism set-up, for example) and in good photography and animation.

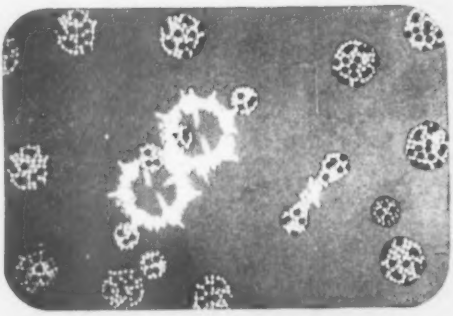
Titles and scenes from films now available are shown here; other films deal with biological effects of radiation, industry and engineering, medical research, diagnosis, and therapy.

United World Films, Inc. (New York 29), will sell prints; the Isotopes Division, AEC (Oak Ridge), and Signal Corps Film Libraries at local Army Area headquarters will lend them.

THE CAMERA FOCUSES ON **THE RADIOISOTOPE**

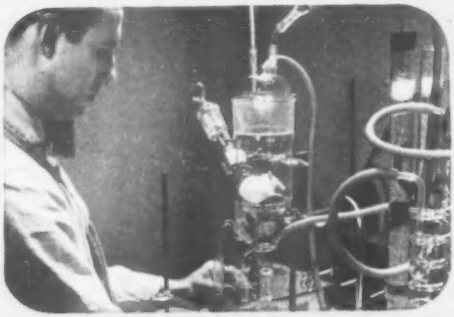


"PRACTICE OF RADIOLOGICAL SAFETY." Equipment is rinsed in a "hot" sink used for contaminated material

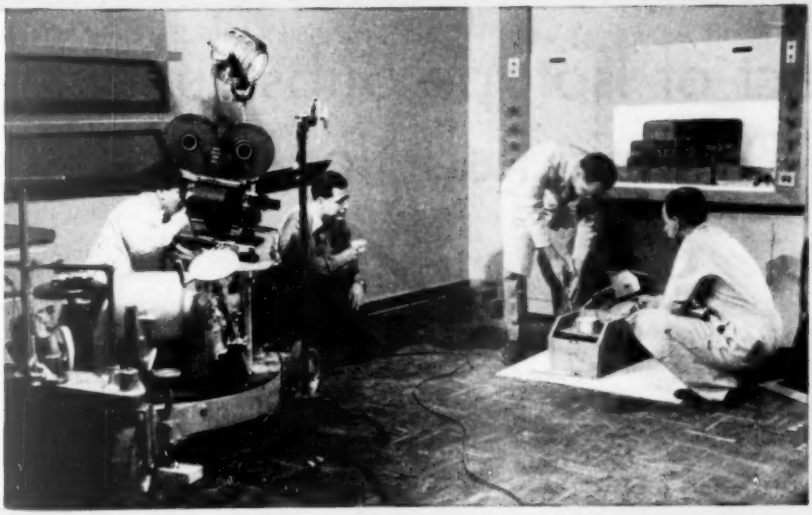


"FUNDAMENTALS OF RADIOACTIVITY." Concept of uranium fission is explained by use of animated pictures

"PRINCIPLES OF RADIOLOGICAL SAFETY." X-ray lab of 1900 era is example of minimum radiation protection



"METHODOLOGY." C^{14}-metabolism experiment with rat in respiration chamber is common and typical tracer study



"PRACTICAL PROCEDURES OF MEASUREMENT." Blood is removed from rabbit to measure P^{32} uptake

"PROPERTIES OF RADIATION." Nature of gamma rays is demonstrated by their penetration of a paper barrier

"AGRICULTURAL RESEARCH." Monitor checks surface radiation from radioactive fertilizer mixed with the soil

"GENERAL SCIENCES." Copper block is plated with radiocopper to study self-diffusion of copper under heat

Effect of Isotope Substitution on Organic Reaction Rates

Studies of C, N, and O isotope effects, mainly undertaken since 1948, necessitate reinterpretation of the quantitative results of some tracer experiments. This review covers recent work on inter- and intra-molecular isotope effects

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IN THE PAST DECADE, many tracer studies involving organic compounds containing isotopes of carbon, nitrogen, and oxygen have been described in the scientific literature. In most of these the assumption was made that the chemical behavior of the molecules bearing the labeled atoms was identical with that of the corresponding unlabeled molecules. Although the effect of deuterium substitution for hydrogen on reaction rates and equilibria had been demonstrated, it was usually felt that no such appreciable effects could be detected as a result of isotopic substitution of the heavier atoms.

Even though isotope substitution was shown to have a small effect on the equilibrium constants of reactions, such as the exchange between hydrogen cyanide and carbon-13-labeled cyanide ions (*1*), larger effects on rate processes were apparently not generally considered to be probable. With the publication of the work of Beeck, Otvos, Stevenson, and Wagner (*2, 3*), and Yankwich and Calvin (*4*), it was demonstrated that the substitution of isotopic carbon atoms could sometimes alter reaction rates enough to cause doubt about the quantitative interpretation of tracer experiments.

Recently many other papers have been published showing qualitatively or quantitatively the effect of C^{13}, C^{14}, N^{15}, and O^{18} isotopes on organic or inorganic reaction rates. In some of these cases, attempts have been made to correlate the empirical rate effects with calculations based on the theory of absolute reaction rates as applied by Bigeleisen, Eyring and others.

In other cases, it has been shown that the alteration of reaction rates by isotopic substitution may be not just a complicating factor in quantitative interpretation of tracer experiments, but also a valuable tool for studying reaction mechanisms (*5, 6, 7*). For example, the carbon isotope effect has been used as an aid in locating the rate-controlling step in a series of organic reactions (*6, 7*). This technique was applied earlier using H^2 (*8*).

Definition of "Isotope Effect"

The term "isotope effect" has been applied in the chemical literature to the effect of isotopic substitution on both the reaction-rate constant and the equilibrium constant, as well as in other senses. In this paper, the term will usually refer to the effect on a reaction rate caused by the substitution of one carbon, nitrogen or oxygen isotope for another.

From the standpoint of correlating isotope effects with theoretically calculated values, there are advantages in being able to express measured and calculated isotope effects quantitatively in terms of rate constants. This correlation appears to be impractical in some cases involving complicated reactions. When quantitative evaluation has been achieved, isotope effects have been variously expressed as k^*/k, $100(k^*/k - 1)$, k/k^*, $100(k - k^*)/k$, etc., where k is the specific reaction rate of the normal molecule and k^* the specific reaction rate of the labeled molecule.

It seems possible that the accumulated effect of a number of isotope effects in a sequence of rate and equilibrium processes of a complex biological system might have a considerable influence on the outcome of a tracer study involving the system. Such a case has not yet been demonstrated experimentally in the tracer uses of carbon, nitrogen, or oxygen isotopes. However, Fry and Calvin (*9*) have discussed the possible multiplication of rate-isotope effects by other isotope effects occurring in equilibria previous to the rate steps.

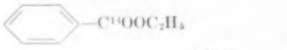
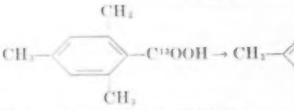
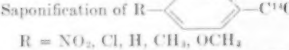
Detection of Isotope Effects

In a simple reaction in which a normal isotope effect is present, i.e., where the heavier molecule reacts faster, the concentration of label in the reacting molecule increases as the reaction proceeds. The concentration of the labeled molecules in the product also increases until both the labeled and unlabeled reactant molecules are completely converted. At the completion of the reaction, the concentration of labeled product equals the initial concentration of labeled reactant.*

The relation of the isotopic com-

* For simplicity this discussion is limited to reactions which do not result in isotopic dilution. If an isotopic dilution accompanies the reaction it must be taken into account.

Experimental and Calculated Values of C^{13}, C^{14}, N^{15}, and O^{18} Isotope Effects

Reaction studied	*Reaction temp. (°C)*	*Determined*	*Experimental value*	*Theoretical value*	*Reference*
Intermolecular Isotope Effects					
Carbon isotopes					
	Room temp.	k_{14}/k_{12}	0.86 ± 0.016	0.877–0.863	(*13, 34*)
	61.2	k_{12}/k_{13}	1.037 ± 0.003	1.038	(*15, 34*)
	92.0		1.032 ± 0.001		(*15, 34*)
	60	k_{12}/k_{13}	1.038 ± 0.001	1.041	(*33, 34*)
Same reaction, carbon-14 labeling	60	k_{12}/k_{14}	1.101 ± 0.007	1.078	(*33, 34*)
$Cl_3C—C^{13}OO^{(-)} \rightarrow Cl_3C—^{(-)} + C^{13}O_2$	70.4	k_{12}/k_{13}	1.0338 ± 0.0007	1.040	(*16, 34*)
$HC^{14}OOH \xrightarrow[H_2SO_4]{} H_2O + C^{14}O$	0	k_{14}/k_{12}	0.8889 ± 0.0052	0.872	(*17; 18,* H. Eyring)
	25		0.9141 ± 0.0058	0.885	
Polymerization of styrene (α-C^{14}) and vinylchloride		k_{14}/k_{12}	0.93		(*35*)
Saponification of  R = NO_2, Cl, H, CH_3, OCH_3	25	k_{14}/k_{12}	0.937 to 0.920		(*14*)
$2C^{13}H_2O + OH^{(-)} \rightarrow C^{13}H_3OH + HC^{18}OO^{(-)}$	60	k_{18}/k_{16}	0.9434 ± 0.0012		(*36*)
$H_2N—C^{13}O—NH_2 + H_2O \xrightarrow{\text{urease}} C^{13}O_2 + 2NH_3$	30	k_{12}/k_{13}	1.010		(*28*)
$H_2N—C^{14}O—NH_2 + H_2O \xrightarrow{\text{urease}} C^{14}O_2 + 2NH_3$	—	k_{12}/k_{14}	$k_{14} > k_{12}$		(*24*)
	28	k_{12}/k_{14}	$k_{12} > k_{14}$		(*37*)
	30	k_{12}/k_{14}	1.032		(*28*)
Absorption of $C^{13}O_2$ in $Ba(OH)_2$		k_{12}/k_{14}	1.014, 1.011		(*38*)
Oxygen isotopes					
Reduction of $H_2O^{16}O^{18}$ by Fe(II), Sn(II), Cr(II), Cu(II)	25	k_{18}/k_{16}	0.932 to 0.95	0.939	(*5*)
Reduction of $H_2O^{16}O^{18}$ by Ti(III)	25	k_{18}/k_{16}	0.994		(*5*)
Oxidation of $H_2O^{16}O^{18}$ by Ce(IV), Cl_2, HOCl	25	k_{18}/k_{16}	0.987–0.992		(*5*)
Intramolecular Isotope Effects					
Malonic acid decarboxylations					
$CH_2(COOH)_2 \xrightarrow{k_1} CO_2 + CH_3COOH$	137	$k_1/2k_2$	1.037 ± 0.002	1.021	(*12*)
				1.035	(*34*)
$C^{13}H_2(COOH)_2 \xrightarrow{k_2} CO_2 + C^{13}H_2COOH$	137	k_4/k_3	1.020	1.0198	(*12*)
$C^{13}OOH \xrightarrow{k_3} C^{13}O_2 + CH_2COOH$	138	$k_1/2k_2$	1.046	1.021	(*29*)
				1.035	(*34*)
CH_2 / $COOH \xrightarrow{k_4} CO_2 + CH_2C^{13}OOH$	138–199	k_4/k_3	1.021–1.024	1.0198	(*29*)
	138	k_4/k_3	1.026–1.027	1.0198	(*32*)
	137–196	$k_1/2k_2$	1.036 ± 0.003	1.021	(*39, 12*)
				1.031–1.035	(*34*)
	137–196	$k_1/(k_3+k_4)$	1.025 ± 0.003		(*39*)
	160	$k_1/(k_3+k_4)$	1.020		(*18,* R. Bernstein)
$CH_2(COOH)_2 \xrightarrow{k_1} CO_2 + CH_3COOH$	>100	k_3/k_2	1.12	1.038	(*4, 11*)
$C^{13}OOH \xrightarrow{k_2} C^{13}O_2 + CH_2COOH$	153–4	k_3/k_2	1.06 ± 0.02	1.038	(*40*)
CH_2 / $COOH \xrightarrow{k_3} CO_2 + CH_2C^{13}OOH$	138	k_3/k_2	1.087 to 1.105	1.038	(*32*)
	154	$k_1/2k_2$	1.065	1.041	(*11, 41*)
	154	$k_3/2k_2$	1.005	1.0026	(*11, 41*)
$CH_2(COOH)_2 \xrightarrow{k_1} CO_2 + CH_2COOH$					
$C^{14}H_2(COOH)_2 \xrightarrow{k_2} CO_2 + CH_2COOH$	154	k_1/k_2	1.077		(*41*)

(Continued on p. 25)

position of the product to the initial isotopic composition of the reactant has been worked out by Bigeleisen (*10*) for several values of k^*/k at all percentages of reaction from 0 to 100. Except in some cases to be discussed under intramolecular isotope effects, no isotope effect can be detected in a reaction which has gone to completion. Hence, errors in tracer experiments due to simple isotope effects can usually be eliminated by carrying all reactions to completion.

Quantitative Evaluation of Effects

Since C^{14}, N^{15}, and O^{18} are not ordinarily available carrier-free, it is obviously impractical to evaluate k^* directly by following the rate of the pure labeled compound, and then to divide by k, the value for the unlabeled compound. Recourse has therefore been had to relating the ratio of the rate constants to some function involving the tracer concentrations in the reactants, the concentrations in the products, or both, and the percent reaction, or time. The general term "tracer concentration" is applied to include both the specific activity in labeling by radioisotopes and isotope concentration in labeling by stable isotopes. It is understood that specific activity is proportional to isotope concentration. In the case of carbon-14, the only radioisotope mentioned here, the half-life is so long that no correction for radioactive decay is necessary.

Intermolecular Isotope Effects

The direct methods involved in studying intermolecular isotope effects exclude studies on reactions of the malonic acid type where a labeled compound can react by several paths.

Tracer concentration of accumulated product vs k^*/k. Stevens and Attree (*11*) derived the expression

$$k^*/k = \ln(1 - rf)/\ln(1 - f)$$

where f = fraction reacted, and r = ratio of the specific activity (C^{14} label) of the accumulated product to the initial specific activity of reactant.

The equation was applied to calculation of k_{14}/k_{12} in the saponification of ethyl benzoate-α-C^{14} with excess base. The specific activity of the benzoic acid produced by 10% reaction was compared with that produced by complete saponification of the ester.

Attempts to apply this method to much faster reactions, such as the saponification (*12*) of ethyl *p*-nitrobenzoate-α-C^{14}, proved unsuccessful. Bothner-By and Bigeleisen (*13*) applied a modification of this method to the decarboxylation of mesitoic acid labeled with C^{13} in the carboxyl group. The equation was transformed so that several such experiments could be combined as a linear plot, the slope of which was taken as a measure of the isotope effect. The equation used was

$$\log\left[1 + \left(\frac{N_x^\circ - N_x}{N_x^\circ}\right)\left(\frac{f}{1-f}\right)\right] = \left(\frac{k_{13}}{k_{12}} - 1\right)\log(1-f)$$

where f = fraction reaction complete, N_x = tracer concentration of carbon dioxide accumulated, and N_x° = tracer concentration of carbon dioxide at complete reaction; $k_{13}/k_{12} - 1$ was measured by the slope of the plot of $\log(1-f)$ versus the left-hand side of the equation for a number of decarboxylations carried to different degrees of completeness.

Bigeleisen and Allen (*14*) have discussed the effect of impurities and assay errors on isotope effects calculated by this method. They have shown that the effect of assay errors on the calculated values of $k^*/k - 1$ increases as the value of f increases, and is particularly great at values of $f > 0.90$. They also have pointed out the greater precision obtainable by mass spectrometric methods with C^{13} as compared with that obtainable by radioassay with C^{14}. This advantage is, of course, partly offset by the approximately two-times-larger isotope effects with C^{14} reactions as compared with those in reactions involving C^{13}. It should also be pointed out that to limit carbon-isotope-effect studies to C^{13} would reduce drastically the number of practical carbon-isotope studies, since isolation of the C^{13}-labeled atom in suitable form for mass spectrometric analysis is often difficult or impossible. With C^{14}, radioassay can be performed on most pure organic compounds, and the specific activity of the compounds can be used in the calculation of isotope effects. With the nitrogen and oxygen isotopes, isotope concentrations must be determined by mass spectrometric measurements. However, C^{13} studies are often preferred because no synthesis of labeled compounds is necessary. The 1% of C^{13} normally present in stock reagents serves as the tracer isotope.

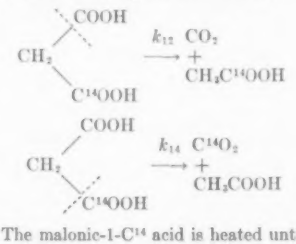
Differential methods applied to first-order reactions. **1.** The plot of the logarithm of the instantaneous specific activity of $C^{14}O$ evolved versus time, for the sulfuric acid dehydration of formic-C^{14} acid (*15*), made possible the determination of $(k_{12} - k_{14})$ from the slope. The C^{14} effect was then calculated by dividing $(k_{12} - k_{14})$ by k_{12}, the kinetically measured first-order rate constant. This gave $100(1 - k_{14}/k_{12})$. Stranks and Harris (*16*) have studied the acidic decomposition of carbonatotetrammine cobaltic complex ion (C^{14} labeled) in a similar way.

2. By plotting the logarithm of the specific activity of remaining malonic-2-C^{14} acid versus time (*12*), it is possible to calculate, from the slope and the kinetically measured rate constant, $100(1 - k_{14}/k_{12})$ for the dry decarboxylation of malonic-2-C^{14} acid at 154°. A somewhat similar method was used to study the Cannizzaro reaction of methanol-C^{14} (*49*).

Other methods. Lindsay, McElcheran, and Thode (*17*) applied to the study of the isotope effect in the decomposition of labeled oxalic acid a simple method which has also been used to evaluate the isotope effects (*12*) in the saponification of a series of ring-substituted ethyl benzoates labeled in the carboxyl groups. The value of k_{14}/k_{12} was calculated as S_0/S_{100}, the ratio of the specific activity of the first acid produced by saponification to the specific activity of the original ester.

Intramolecular Isotope Effects

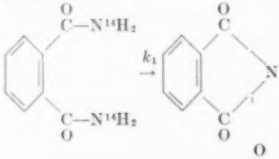
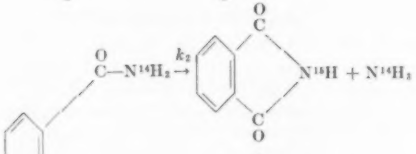
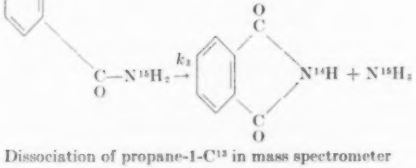
The decarboxylation of carboxyl-labeled malonic acid is a typical example of the intramolecular type, which is often more easily studied than the intermolecular type just discussed. With C^{14} labeling the reactions are



The malonic-1-C^{14} acid is heated until completely decomposed to acetic acid and carbon dioxide. The two products are collected and their specific activities are determined. The relationship

$$k_{14}/k_{12} = S_{CO_2}/S_{HAc}$$

Isotope Effect Values (continued)

Reaction studied		Reaction temp. (°C)	Determined	Experimental value	Theoretical value	Reference
$C^{14}OOH \xrightarrow{k_1} C^{14}O_2 + RCH_2C_2OH$ RCH $COOH \xrightarrow{k_2} CO_2 + RCH_2C^{14}O_2H$	R = Br	100	k_2/k_1	1.4	1.038	(4)
	R = Br	138	k_2/k_1	>1.1	1.038	(32)
	R = α-naphthyl	73–95	k_2/k_1	1.097 ± 0.008	1.038–1.14	(20, 18)
	R = α-naphthyl	163	k_2/k_1	1.076 ± 0.004	1.038–1.14	(20, 18)
	R = phenyl	73–95	k_2/k_1	1.132 ± 0.015	1.038–1.14	(20, 18)
	R = phenyl	163	k_2/k_1	1.088 ± 0.015	1.038–1.14	(20, 18)
Oxalic acid decompositions in H_2SO_4						
$(COOH)_2 \xrightarrow{k_1} CO_2 + CO + H_2O$						
$C^{13}OOH \xrightarrow{k_2} C^{13}O_2 + CO + H_2O$		100	k_2/k_3	1.032		(19)
		100	$k_1(k_2 + k_3)$	1.034		(19)
$COOH \xrightarrow{k_3} CO_2 + C^{13}O + H_2O$		103	k_2/k_3	1.027 ± 0.0022	1.0227–1.031	(9, 18)
		80	k_2/k_3	1.033 ± 0.0015	1.0227–1.034	(9, 18)
Same reaction of oxalic-C^{14} acid		103	k_2/k_3	1.055 ± 0.0030	1.0435–1.060	(9, 18)
		80	k_2/k_3	1.067 ± 0.0035	1.0435–1.063	(9, 18)
Other reactions						
		180	k_2/k_3	1.014 ± 0.001	1.016	(7, 26)
			$k_1/2k_2$	0.994 ± 0.001	>0.97	(7, 26)
			k_3/k_2	1.2		
Dissociation of propane-1-C^{13} in mass spectrometer						
$H_3C—CH_2—CH_3 \xrightarrow{k_1}$			$k_1/2k_2$	1.12 ± 0.1	1.030(500° K)	(2, 42)
$H_3C^{13}—CH_2—CH_3 \xrightarrow{k_2}$			$k_1/2k_3$	0.93 ± 0.002	1.010(500° K)	
$H_3C^{13}—CH_2—CH_2 \xrightarrow{k_3}$			k_2/k_3	1.2		
Thermal cracking of propane-1-C^{13}		500–550	k_2/k_3	1.08		(3)
$O=C^{14}—C_6H_5 + NaOH \xrightarrow{k_{14}} (C_6H_5)_2C(OH)C^{14}OONa$			k_{12}/k_{14}	1.11 ± 0.01		(43)
$O=C—C_6H_5 + NaOH \xrightarrow{k_{12}} (C_6H_5)_2C^{14}(OH)COONa$						
$O=C\begin{matrix} \diagup C^{14}H_3 + 2NaOH + I_2 \xrightarrow{k_{14}} C^{14}H_3I + CH_3COONa + NaI + H_2O \\ \diagdown CH_3 + 2NaOH + I_2 \xrightarrow{k_{12}} CH_3I + C^{14}H_3COONa + NaI + H_2O \end{matrix}$			k_{14}/k_{12}	1.03 to 1.06		(25)
$Ni(CO)_4 \xrightarrow{k_1} Ni(CO)_3 + CO$						
$Ni\begin{matrix} \diagup C^{13}O \xrightarrow{k_2} Ni(CO)_3 + C^{13}O \\ \diagdown (CO)_3 \xrightarrow{k_3} Ni\begin{matrix} \diagup (C^{13}O) + CO \\ \diagdown (CO)_2 \end{matrix} \end{matrix}$			$k_1/(k_2 + k_3)$	1.034 ± 0.004		(18)
			$k_1/4k_3$	1.059		(18)

(Continued on p. 27)

where S refers to specific activity, is apparent since, at complete reaction, the number of moles of each labeled product formed is proportional to its specific rate of formation. During the reaction, the relationships between the concentrations of label in the reactant and products are more complex, although they have been completely worked out (*18, 19*).

Intramolecular isotope effects in the reactions of unsymmetrically labeled symmetrical compounds have also been evaluated (*4, 20*) by measuring the isotope concentrations in the labeled reactant and in a labeled product after complete reaction.

In the cases of the malonic-1-C^{14} acid decarboxylation and the malonic-1-C^{13} acid decarboxylation, both types of studies have been carried out, i.e., studies involving the unsymmetrically-labeled symmetrical molecules (intramolecular isotope effects) and studies involving the relative decarboxylation rates of the labeled and unlabeled molecules (intermolecular isotope effects). In general, the methods of calculation have involved combinations of the above methods, i.e., studies of the tracer concentrations in the reactants and products at measured percentages of reaction.

Theoretical Calculations

Outstanding among theoretical treatments of the isotope effect on reaction rates have been those of Bigeleisen (*21*), who calculated the ratio of rates of reaction of isotopically-labeled compounds to be

$$\frac{k}{k'} = \left(\frac{\mu'}{\mu}\right)^{1/2}\left[1 + \sum_{1}^{3N-6} G(u_i)\Delta u_i - \sum_{j}^{3N'-6} G(u_j\ddagger)\Delta u_j\ddagger\right]$$

where the symmetry is the same in the labeled and unlabeled molecules. Here μ is the reduced mass, $m_1m_2/(m_1 + m_2)$, calculated using m_1 and m_2 as the masses of the atoms involved in the bond being broken or formed. The term in brackets is said to express the zero-point energy effect; $G(u)$ is the function (*22*) $1/2 - 1/u + 1/(e^u - 1)$, where u is $h\omega/kt$, and $\Delta u = (hc/kt)\Delta\omega$; $\Delta\omega$ is the effect of isotope substitution on a given vibrational frequency in the molecule. Starred terms refer to the activated complex. The expression was derived on the basis of the theory of absolute reaction rates with the assumption that the effect of the isotope mass on potential energy is negligible. It states that there are two principal ways in which varying the isotope mass changes the reaction rate: (1) the zero-point energy effect, which expresses the effect of mass on the equilibrium between the reactants and the activated complex, and (2) the reduced-mass effect, which expresses the effect on the frequency of rupturing the critical bonds of the activated complex. It may be noted here that factor (1) is stated to reduce to approximately unity for isotope effects of the intramolecular type (*7, 18*) such as that in the decarboxylation of malonic-1-C^{14} because the same molecule decomposes in two ways; in these cases the theoretical value of the isotope effect is simply calculated as a reduced-mass effect which is not dependent on temperature.

Using Bigeleisen's equation, Pitzer (*23*), Bigeleisen and others have compared experimental and theoretical values of a number of isotope effects. Only in some cases can the agreement be said to be good. Usually agreement seems to be poorer with C^{14} than with the other isotope effects. Among the reasons for poor agreement may be the fact that in many of the molecules whose rates of reaction were studied the vibrational frequencies have not yet been completely worked out from spectra—rather crude approximations have sometimes been necessary. Furthermore, the decision as to what frequencies are of critical importance in calculations of the zero-point energy effect has in certain cases been difficult. Thus Pitzer (*23*) and Bigeleisen (*18*) developed different models for their calculations in the case of the decarboxylation of malonic-1-C^{14} acid. This calculation refers to that isotope effect which is a comparison between the rates of two molecules, the labeled and the unlabeled. For this calculation, Pitzer (*23*) obtained $k/k' = 1.14$ as a limiting value, whereas Bigeleisen (*18*) obtained 1.04. It must be admitted, however, that different workers have found quite different experimental values as well for some isotope effects.

In agreement with experiment, the theory would predict (*21*) that isotope effects for carbon, nitrogen and oxygen isotopes should become smaller at higher temperatures. It also would predict that the largest normal isotope effects should occur in those reactions in which the labeled atom is tightly bound in the reacting molecule, and nearly free in the activated complex. Bigeleisen (*10*) has estimated the maximum possible values of isotope effects at 25° C for a number of isotopes by assuming the labeled atoms to be free in the activated complexes. Several estimated reaction rate ratios were: C^{12} to C^{14}, 1.5; N^{14} to N^{15}, 1.14; and O^{16} to O^{18}, 1.19. Bigeleisen (*21*) has also predicted on the basis of the theory that in no case involving a tracer atom heavier than hydrogen can the molecule bearing the heavy isotope react faster. Although this has usually been found to be the case, several "reverse-isotope effects" have been reported (*24, 25, 26*). At least one of these special cases has been plausibly explained (*7*), and another (*24, 27, 28*) has been discredited on the basis of new experimental data.

Tabulation of Isotope Effects

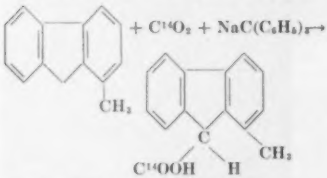
An attempt has been made in the table to list the published carbon, nitrogen, and oxygen isotope effects in organic reactions. Where possible, the isotope-effect values have been stated in terms of the appropriate rate constants, as they were originally published. The corresponding theoretically calculated values have also been listed.

A number of other aspects of isotope effects on reaction rates of organic compounds are perhaps worth mentioning. It seems clear from the results of several studies (*13, 15, 20*) that the greatest isotope effects are to be expected at low temperatures. In cases of intramolecular isotope effects, however, the degree of fractionation had been found to be temperature independent (*29*) in accord with theory.

Further experimental work is necessary to clear up the picture with regard to reverse isotope effects, the existence of rather large fractionation factors in certain equilibria (*30*) and the effect of reaction conditions such as choice of solvent concentration or catalyst on the extent of isotope fractionation.

Very few isotope effects have been demonstrated in simple addition reactions. Certain work involving deuterium (*31*) suggests that greater isotope effects are to be expected with slower reactions, but further research is needed to determine whether such

Isotope Effect Values (continued)

Reaction studied	Reaction temp. (°C)	Experimental value	Ref.
Isotope Effects Reported Qualitatively, etc.			
	−80	$k_{12} > k_{14}$	(27)
$2HgOOC^{14}H \rightarrow 2Hg + HC^{14}OOH + C^{14}O_2$	70–75	$k_{12} > k_{14}$	(14)
$[Co(NH_3)_4C^{12}O_3]^{\oplus} + C^{14}O_3^{\ominus} \overset{K}{\rightleftharpoons} [Co(NH_3)_4C^{14}O_3]^{\oplus} + C^{12}O_3^{\ominus}$	0	$K = 0.875 \pm 0.002$	(18)
	20	$K = 0.89$	(30)
	30	$K = 0.900 \pm 0.004$	(18)
$CH_3—C^{14}OOH + 4O \xrightarrow[\text{Oxidation}]{\text{Van Slyke}} 2C^{14}O_2 + 2H_2O$	100	$k_{12} > k_{14}$ Theoretical value = $1.075(k_{12}/k_{14})$	(44, 18)
$C^{14}O_2$ absorption in photosynthesis		$k_{12} > k_{14}$	(45)
Combustion of C^{14}-labeled xanthydrol ureide		$k_{12} > k_{14}$(?)	(46)
Reactions in which no isotope effect was found			
Pyrolysis of barium adipate (normal C^{13} concentration)	350		(6)
Benzoin condensation and Cannizzaro Reaction(C^{14})			(43)
Decarboxylation of *o*-aminobenzoic acid-α-C^{14}			(47)
Acid hydrolysis of *t*-butylbenzoate-α-C^{14}			(47)
Pyrolysis lithium acetate-1-C^{14}	490		(48)
Absorption $C^{14}O_2$ in alkali and benzylamine	25		(49)
Bromination of styrene-α-C^{14} and styrene-β-C^{14}	−70		(14)
Hydration of diphenylacetylene-1-C^{14}			(50)
Diels-Alder reaction of 2,3-dimethyl-butadiene with β-nitrostyrene-α-C^{14}	80–100		(14)
Acidic decomposition of C^{14}-labeled carbon-atotetrammine cobaltic complex ion	25		(18)

is the case with isotopes of carbon, nitrogen and oxygen. Ratios of the C^{14} isotope effect to the C^{13} isotope effect (for the same reaction) have been observed (*32*, *33*) as high as 3 to 1 and 4 to 1. The theory, according to Bigeleisen (*34*), predicts a ratio of approximately 2 to 1. This disagreement of theory and experiment remains unexplained.

Cahill and Taube (*5*) have discussed the usefulness of isotope fractionation studies as applied to investigation of reaction mechanisms. Undoubtedly this tool will find frequent application in the future. Bernstein (*16*) has discussed the use of rate isotope effects as means of separating carbon isotopes.

It is to be hoped that as more results of isotope effect studies accumulate, some empirical working rules can be formulated to predict in what organic reactions and to what extent isotope fractionations can be expected to occur. Such rules would be of particular value to many users of tracer techniques who do not care to use the theory.

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A Converted Electron Accelerator to Produce 2.3-Mev Positive Ions

FIG. 1. Positive-ion accelerator

A new terminal, tube, energy control system, and pumping equipment were needed to convert an electrostatic electron accelerator. The r-f ion source can produce beams of protons, deuterons, tritons, He-3, and He-4

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THERE IS A growing need for positive-ion accelerators capable of consistent production of high beam currents of charged particles.

The accelerator shown in Fig. 1 produces a 100-μa beam of magnetically analyzed protons up to 2.3 Mev. The beam can be focused into a spot 1 mm in diameter. The beam energy is regulated by a slit and corona system to ± 3 kev. An r-f ion source is used to produce beams of deuterons, tritons, He^3, He^4, or protons.

The basic electrostatic accelerator was designed and constructed by the High Voltage Laboratory of the Massachusetts Institute of Technology. This particular accelerator was one of the five described by Buechner, Van de Graaff, *et al.* (*1*). It was delivered to the Navy in 1945, and later purchased by the Oak Ridge National Laboratory. It was installed and operated as an electron generator with the original equipment. Subsequent to that time it has been converted and operated as a positive-ion generator.

A 3-ft extension has been added to the original steel tank to allow for the increased height of the column, necessitated by the added terminal equipment. The generating voltmeter is mounted in the top of the tank for a voltage monitor. The insulating medium is CO_2 at 200 psig instead of air; however, the gas system is the original one consisting of a storage tank, Kinney pump, cold trap, compressor, and activated alumina dryer.*

The equipotential planes, belt spacers, drive motor, and fabreeka belts are unchanged. The upper belt pulley has been rebuilt to house two 400 volt-ampere, 180 cycle, generators to supply power to the terminal. Five Lucite rods run the length of the column to turn Variacs in the terminal for various voltage controls. The bottom end of these rods are connected through 10:1 gear boxes to selsyn receivers which in turn are controlled by selsyn transmitters at the control desk. With this arrangement, a telemetering system

* Activated alumina was found to be better for drying CO_2 than was silica gel.

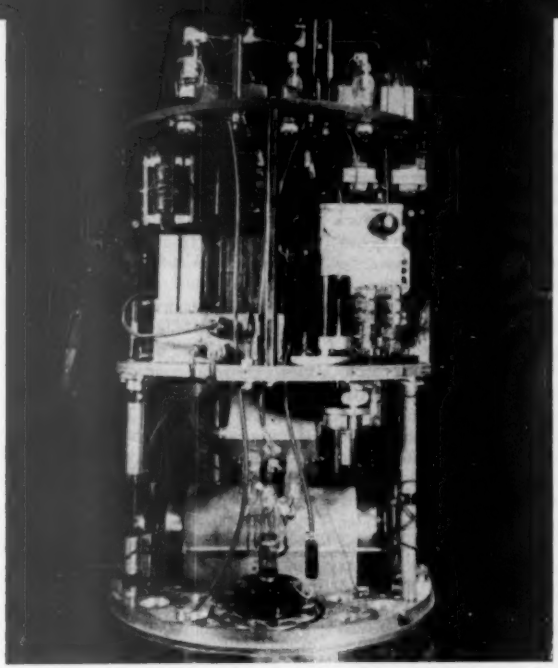

FIG. 2. High-voltage terminal of the accelerator showing the r-f ion source

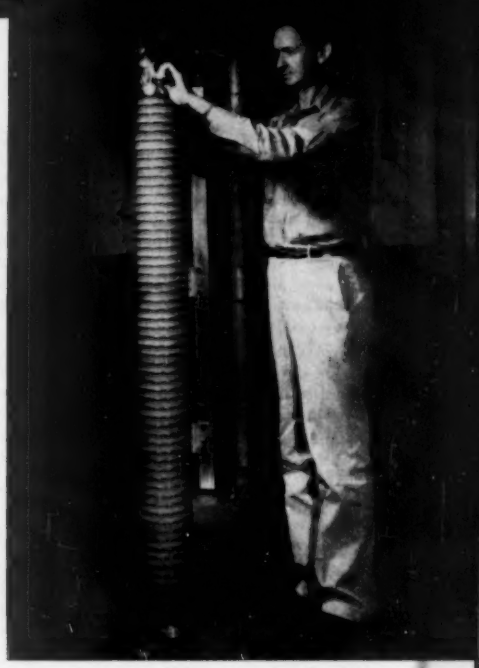

FIG. 3. Positive-ion electrostatic accelerator tube

for monitoring the controls in the terminal was found unnecessary since the selsyn dials on the front panel could be calibrated.

Charge is carried both up and down on the belt. Because of the limited space at the base of the column, only one spray comb was installed to spray on positive charge and take off negative charge. There are two spray combs at the terminal, one for taking off the positive charge and the other for spraying on the negative charge.

The bottom spray comb was modeled after the ones used by the High Voltage Engineering Corp. It consists of 10 groups of 25 needles, each group being connected to the high-voltage bus through a 50-megohm resistor. The entire spray system has proved to be very satisfactory, giving long belt life at 300 μa of charging current.

As a safety precaution for the accelerator tube, spark gaps were mounted between each equipotential plane, spaced such that they would break down with 50 kv across any gap. The gaps were painted with radioactive Cs^{137} to give faster breakdown.

Terminal Equipment

Figure 2 is a photograph of the high-voltage terminal of the accelerator. The r-f ion source, designed by Moak, Reese and Good (*2*), is shown at the bottom. It is capable of supplying up to 300 μa of hydrogen ions, most of which are of mass one. It was found that an ion source with a canal 0.040 in. in diameter and ½ in. long was most suitable for this accelerator because of the low gas consumption.

Immediately behind and above the ion source bottle is a 100-watt, 90-megacycle oscillator used to excite the ion source. The oscillator is coupled to the ion source by two r-f shoes slipped over the bottle. The field coil is shown at the base of the ion source. Preliminary experiments show that the field coil can be replaced by permanent magnets if lower power consumption is required in the terminal.

The upper tiers house a 5-kv positive power supply for the ion source probe, a 15-kv negative supply for the accelerator tube lens, the power supply for the oscillator, and a 90-volt, d-c, supply for the ion source field coil. The other side of the terminal from that shown houses a 30-kv negative supply for the negative belt charging, the gas bottles, and the Variacs.

Deuterium or hydrogen can enter the ion source through a palladium leak.

Accelerator Tube

Since the objective was a proton current of 100 μa at 2 Mev, considerable time was spent in the design of the accelerator tube. It is generally true that the capabilities of a positive ion Van de Graaff are limited by its tube. It was thought that this would be particularly true here since the proton current would be very much higher than customary. The tube described here, however, has more than met expectations.

Figure 3 shows the assembled tube which consists of 56 active sections and 4 extra sections, with a total length of 60 in., inside diameter of 3 in., and outside diameter of 6½ in. The glass is No. 707 Pyrex, and the electrodes are No. 347 stainless steel. Figure 4 shows a cross section of the tube.

Compromise had to be made between pumping speed, shielding factor (the degree to which the glass is shielded from the beam), and minimum spacing between the electrodes. Since the tube might occasionally be used for accelerating electrons, this latter point had to receive more special consideration. Compromise was effected as follows:

1. A 3-in. bore gave a pumping speed of 100 l/sec for H_2. The ion source discharges 1.3 μ-l/sec into the upper end of the tube, and this results in an increase in pressure of 0.013 μ between the bottom and the top.

2. The shielding factor adopted was

87%. This is a measure of the degree of geometrical shielding afforded by the electrodes between an axial beam and the inner glass walls. This factor proved to be entirely adequate, as no drifting of the beam occurs in operation.

3. The minimum permissible spacing between the closest points of successive electrodes will be fixed by the required negative potential, as this will make the lip of the upper electrode negative with respect to the inner plane surface of the following one. It is under these conditions that cold emission is most likely to occur. The spacing was chosen as 0.25 in., which for negative operation gave a ratio of 6.0 for surface gradient in kilovolts per centimeter to section voltage in kilovolts. Under these conditions it was found that a single section would withstand at least 50 kv for negative polarity and 100 kv for positive polarity. A total operating potential of 2 Mv requires that each section operate at 36 kv.

Voltage division down the column is accomplished by a string of 112 (2 per section) 250-megohm, type 80X resistors, manufactured by S. S. White. Although quite costly ($18.00 each), they were found to be fully justified in view of their very great simplicity and superiority to corona points.

The lens was designed by R. O. Case,* and consists of a single element whose potential can be varied from 0 to −15 kv with respect to the terminal. The potential is led into the lens via the second electrode from the top of the tube. The electrodes at the bottom are connected to a d-c source, 5 kv negative with respect to ground. This provides a potential barrier to prevent electrons generated by positive ion collisions with residual gas molecules below the bottom of the tube from being drawn up into the tube and being accelerated. When operating at high currents this was found to reduce slightly the amount of bremsstrahlung coming from the top of the tube. For a terminal potential of 2 Mv, −5 kilovolts on these two sections is sufficient to establish a barrier of −3.9 kv on the axis of the tube. A few hundred volts is sufficient to keep out the electrons.

The two materials most commonly used for electrodes are stainless steel and aluminum. Stainless steel suffers from the disadvantage of producing more bremsstrahlung from the secondary electrons which are always present to some degree in high-voltage tubes such as this. However, it is practically free from damage due to sparking, and was chosen for this reason.

The shape of the electrodes is thought to be quite important, and a rather careful analysis was made of the electrostatic field inside the tube. The field was determined experimentally first by the use of an electrolytic tank; however, such a method suffers from lack of resolution in regions close to the electrode surfaces. Since these regions are by far the most important, the accuracy here was improved by solving Laplace's equation for these regions by successive approximations (sometimes referred to as the Liebnitz network procedure) (*3*). Figure 5 shows the flux plot obtained in this manner. The electrodes are so shaped that secondary emission of electrons is inhibited in the region of the inner rim.

The curve shown in the figure is the trajectory of an electron emitted with not over a few volts energy from the plane surface of the electrode at a point just above the region of the retarding field. It is seen that this electron will travel the length of the tube without striking another electrode. This point should be noted carefully as it is thought to be important in good design, since if such secondary electrons struck other electrodes on their journey up the tube, electron multiplication could result. This process can lead to instability and tube breakdown, in accordance with the theory of Trump and Van de Graaff (*4*).

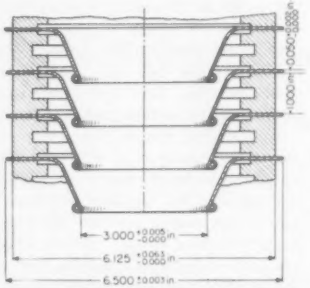

FIG. 4. Cross section of positive-ion accelerator tube

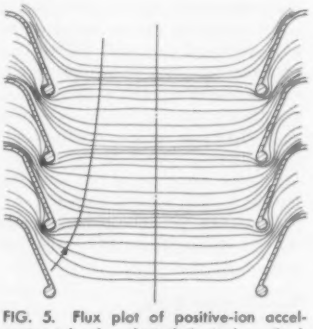

FIG. 5. Flux plot of positive-ion accelerator tube by electrolytic tank method. Curve shows an electron trajectory

The trajectory shown was obtained by the step by step solution of the equations of motion of an electron in an electrostatic field. The shape of the trajectory is determined almost entirely by the field near the electrode which emits the particle.

As a check on the importance of this phenomenon, a tube was constructed identical in all respects to the one described here except that the electrodes were simply flat sheets with a 3-in. bore. This tube would not operate above 2 Mv, whereas the proton tube was operated up to 2.6 Mv without sparking. Tests at higher voltages could not be carried out because of the sparking between the high-voltage terminal and the pressure tank. There was no indication that the presence of a beam affected the insulating ability of the tube.

Pumping System

The pumping system was chosen primarily for its ability to handle hydrogen. The pumping speed of an oil diffusion pump for hydrogen may be as low as 10% of the pumping speed for air due to the back diffusion of the light molecules through the vapor stream. Increasing the density of the vapor stream, by application of more heat for example, greatly reduces this back diffusion and causes the pump to have a greater pumping speed (due to the higher thermal velocity of the light molecules) for hydrogen than for air. This also helps to keep the fore-pressure low. High vapor density in the jets requires better baffling to keep the oil out of the vacuum system.

The choice made was an MCF-300 diffusion pump backed with a VKB-8 booster pump, both manufactured by Distillation Products Industries. The

* Now with North American Aviation Corp.

VKB-8 was in turn backed by a Welch rotary oil pump, type 1405B. This combination is well matched and pumps at 330 l/sec for hydrogen, and 150 l/sec for air, both at 0.3 μ.

A baffle was installed just above the diffusion pump. The plates were kept at a temperature of $-60°$ C by means of a simple two-stage refrigeration system using Freon 22. The baffle is quite effective, designed so that the minimum number of collisions between a molecule traversing the baffle and a refrigerated surface is four. With this system, the pressure with no beam is 0.002 μ, and with a beam will not exceed 0.007 μ, at the base of the accelerator tube.

In addition to the baffle, a large appendix-type cold trap was connected to the system with a high-vacuum valve interposed. In normal operation it is not necessary to use the cold trap but its use may be desirable when the absolute minimum of oil and other condensable vapors is required. There is some evidence that a further gain can accrue by the use of a mercury diffusion pump in place of the oil pump.

Energy Control

A block diagram of a widely used energy control system is shown in Fig. 6. This system functions by intercepting a small fraction of the beam current on two tantalum plates located between the analyzer magnet and the target, forming a slit for the beam to pass through. These insulated plates are returned to ground through separate resistors.

A beam of charged particles of a given energy is analyzed by the highly stabilized magnet and passed through the slit system. The spacing between the plates is adjusted to give the desired energy resolution. As the beam energy changes, more beam will strike one plate depending on which way the correction is needed, causing a difference in potential between the two plates. This signal is then amplified by the d-c amplifier and applied to the grid of a triode-connected 4-125A vacuum tube. This tube, in series with a set of adjustable-position corona points, forms a shunt path between the high-voltage terminal and ground. This system is capable of controlling the terminal voltage over a range of 40 kv due to the high impedance of the Van de Graaff machine. This constitutes the fast response, limited range portion of the control system.

To make the system responsive to large, slow variations such as changes in beam current, or to manual adjustment of the analyzer magnet flux, a voltage proportional to the average corona current is developed across a small resistor between the 4-125A filament and ground. This voltage, of the order of 2 mv, is balanced against an equal voltage obtained from a voltage divider and a 1½-volt battery. Any unbalance voltage developed here is fed into a Brown Electronik amplifier whose motor drives a Variac which in turn changes the belt charging current of the machine in a direction which restores the original average corona current. Thus, to run the high-voltage terminal through its entire range, it is necessary only to run the analyzer-magnet flux through an appropriate range; the regulated terminal voltage will follow, holding the beam on target.

One other manual adjustment must be made from time to time as the terminal voltage changes through large limits. This is the mechanical position of the corona points with respect to the terminal, and the position must be so related to the actual terminal voltage as to cause the 4-125A to operate near the center of its control range. This center is indicated by a zero-center meter on the panel of the amplifier.

Magnetic Analyzer

The magnetic analyzer consists of a brass vacuum chamber, 1 in. thick, mounted between the pole faces of an Armco-iron magnet. There is an exit port for a straight-through beam, and one at 22 degrees from the vertical.

Power to the magnet coils is supplied by a 500-volt, 500-ma electronically regulated power supply. The magnetic field is continuously variable up to 6,000 gauss and is measured to ± 1 gauss by a Model F-6 nuclear fluxmeter made by Varian Associates. The probe of the fluxmeter is mounted between the pole faces adjacent to the analyzer chamber.

The analyzer chamber houses tantalum beam-defining slits used in conjunction with the energy control system. A glass section containing both a quartz and tantalum beam viewer is mounted just above the chamber. The magnet and analyzing chamber are to be replaced with a 90-degree bender to provide better energy resolution and a horizontal beam.

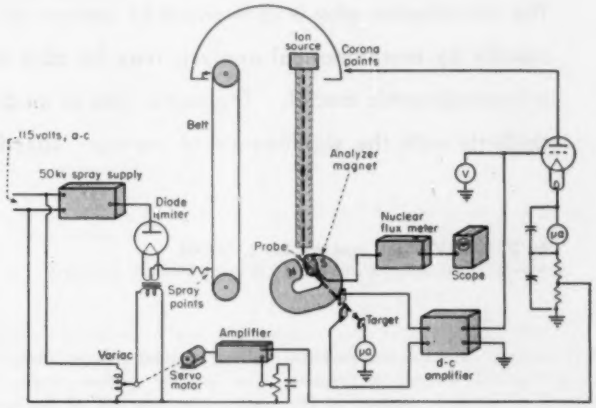

FIG. 6. Energy control system of positive-ion accelerator

* * *

The authors wish to express their appreciation to Dr. W. M. Good; to R. W. Bennett for the design of the excellent high-vacuum valves and many other of the intricate mechanical accessories; to R. F. King for the design of the electronic equipment; to J. D. Kington for carrying out many of the computations in the accelerator tube analysis; and to H. Reese, Jr. for the operation and maintenance of the accelerator.

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A Hydrodynamic Model of Isotope Distribution in Living Organisms

The investigator who is confronted by isotope data that cannot be interpreted readily by mathematical analysis may be able to illustrate his problem with a hydrodynamic model. The same kind of model can be helpful in acquainting students with the significance of various "specific activity-time" curves

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RADIOACTIVE and stable isotopes have made it possible to study the dynamic aspects of metabolic systems in the intact animal without producing gross alterations in its steady state.

In the study of phosphorus metabolism, early workers administered radioactive phosphate to animals and then measured the distribution of radioactivity in various tissue constituents. This type of study yields a great deal of information on the fate of administered phosphate, but it is only of limited value in the study of endogenous phosphate metabolism. To study the latter, one has to know more about the isotope dilution that takes place between the time of administration of the labeled phosphate and the time of its incorporation into the compound under study. As a measure of isotope dilution, Hevesy introduced the measurement of "specific activity" of labeled phosphate, or the ratio of the amount of radioactive phosphate to that of the total phosphate.

When labeled inorganic phosphate is injected intravenously, it is diluted immediately by circulating plasma phosphate and subsequently by extracellular and intracellular phosphates. This stepwise dilution results in a continuous reduction of the specific activity of the plasma phosphate.

Eventually the specific activity of this phosphate will be reduced even further by the exchange with bone phosphate, by the turnover of intracellular phosphate esters, and by the ingestion of unlabeled phosphate. Similarly, the intracellular organic phosphates will undergo continuous changes of specific activity. The extent of these changes depends on the turnover rates of the phosphate esters and their precursors.

It is thus evident that the amount of radioactivity present in one of the phosphorus fractions of a given tissue is not only the result of the metabolic activity of that fraction, but depends greatly upon both the specific activity of the phosphorus-containing precursors and the time interval which has elapsed since the injection of P^{32}. Most workers have recognized that the measurement of the specific activity of a compound is not sufficient for the determination of its turnover rate, but that specific activity-vs-time relations of the compound and of its precursors are necessary for this purpose.

It is possible to derive quantitative relationships between the specific activities of metabolites and their turnover rates, making certain assumptions about the homogeneity of mixing, and the manner in which the labeled materials break down. This type of analysis can be applied by the extensive use of mathematics, but it becomes very difficult as the complexity of the situation increases. Since the mere application of formulas derived by others is a poor substitute for the understanding of factors that produce changes in the isotope distribution, we have explored other avenues of approach to this problem.

Let us examine the metabolism of liver and plasma phospholipides. In 1943, Fishler *et al.*, showed that in the fasting dog the liver is the only source of plasma phospholipides (*1*). Three years later, Entenman *et al.* found that the liver is also the organ to which most of the plasma phospholipides return (*2*).

On the basis of these observations, the metabolic pattern of liver and

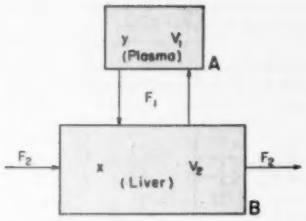

FIG. 1. Theoretical plasma-liver phospholipide system (see text at right)

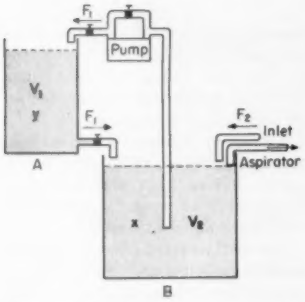

FIG. 2. Hydrodynamic model used to study isotope distribution

plasma phospholipides can be diagrammed as shown in Fig. 1. The following expression for the specific activity of plasma phospholipides after a single intravenous injection of labeled phospholipide can be derived from Fig. 1 and the text accompanying it:

$$y/V_1 = C_1e^{\alpha t} + C_2e^{\beta t}$$

From Eqs. 8 and 9, the two turnover rates F_1 and F_2 can be calculated if the total amount of phospholipide in plasma and liver (V_1 and V_2) and the slopes α and β are known.

A model is shown in Fig. 2 that can be made to portray the relationship between turnover rates, compartment size, and isotope concentration curves. This model consists of two cylindrical jars, one placed at a higher level than the other. Water flows by gravity from A into B, whereas water is pumped back from B into A. The pump is fitted with an outlet valve and a by-pass for regulating its output without changing the speed of the motor. The level of water in jar B is maintained by supplying fresh water, and removing the excess with an aspirator pump. The contents of

IN THE SYSTEM SHOWN IN FIG. 1:

V_1 = amount of phospholipide in the plasma compartment (A)
y = amount of radioactive phospholipide in the plasma at any time t
F_1 = rate of exchange of phospholipide between plasma and liver
V_2 = amount of phospholipide in the liver compartment (B)
x = amount of radioactive phospholipide in the liver at any time t
F_2 = rate of synthesis of new phospholipide from nonlabeled precursors which equals rate of breakdown of labeled phospholipide in the liver

IT IS ASSUMED THAT: (a) a steady state exists where F_1, F_2, V_1 and V_2 remain constant; (b) the radioactive phospholipide is completely mixed with the nonradioactive phospholipide, so that random breakdown and exchange take place; and (c) all liver phospholipides exchange with plasma. (In the dog, the latter is not true since the cephalins do not leave the liver cell.)

One can write:

$$\frac{dy}{dt} = \frac{x}{V_2}F_1 - \frac{y}{V_1}F_1 \tag{1}$$

$$\frac{dx}{dt} = \frac{y}{V_1}F_1 - \frac{x}{V_2}F_1 - \frac{x}{V_2}F_2 = -\frac{dy}{dt} - \frac{x}{V_2}F_2 \tag{2}$$

When $\frac{F_1}{V_2} = a$, $\frac{F_1}{V_1} = b$, and $\frac{F_2}{V_2} = c$,

then $\frac{dy}{dt} = ax - by$, and $\frac{dx}{dt} = -\frac{dy}{dt} - cx$

or, writing d/dt as D, $(D + b)y - ax = 0$ and $Dy + (D + c)x = 0$, which can be rewritten

$$\begin{array}{rl} (D+b)(D+c)y - a(D+c)x = 0 & \\ aDy \qquad\quad + a(D+c)x = 0 & \\ \hline [D^2 + (a+b+c)D + bc]y = 0 & \text{add} \end{array}$$

This is a second-order linear differential equation with constant coefficients which is solved by standard methods (5) to obtain

$$\frac{y}{V_1} = C_1e^{\alpha t} + C_2e^{\beta t} \tag{3}$$

for $\alpha \neq \beta$, where

$$\alpha = \frac{-\left(\frac{F_1}{V_2} + \frac{F_1}{V_1} + \frac{F_2}{V_2}\right) - \sqrt{\left(\frac{F_1}{V_2} + \frac{F_1}{V_1} + \frac{F_2}{V_2}\right)^2 - 4\frac{F_1F_2}{V_1V_2}}}{2} \tag{4}$$

$$\beta = \frac{-\left(\frac{F_1}{V_2} + \frac{F_1}{V_1} + \frac{F_2}{V_2}\right) + \sqrt{\left(\frac{F_1}{V_2} + \frac{F_1}{V_1} + \frac{F_2}{V_2}\right)^2 - 4\frac{F_1F_2}{V_1V_2}}}{2} \tag{5}$$

$$C_1 = \frac{y_0(\beta V_1 + F_1)}{V_1^2(\beta - \alpha)} \qquad y_0 = y \text{ at } t = 0 \tag{6}$$

$$C_2 = \frac{y_0(\alpha V_1 + F_1)}{(\alpha - \beta)V_1^2} \tag{7}$$

or

$$F_1 = \frac{-(\alpha+\beta) \pm \sqrt{(\alpha+\beta)^2 - 4\alpha\beta V_1\left(\frac{1}{V_1} + \frac{1}{V_2}\right)}}{2\left(\frac{1}{V_1} + \frac{1}{V_2}\right)} \tag{8}$$

$$F_2 = \frac{-V_2(\alpha+\beta) \pm \sqrt{(\alpha+\beta)^2V_2^2 - 4\alpha\beta(V_1+V_2)V_2}}{2} \tag{9}$$

When the specific activity y/V_1 is plotted against time on semilogarithmic paper, the numerical values of the slopes α and β (see Fig. 3) are easily obtained from the following relation:

$$\alpha \text{ (or } \beta) = \frac{0.69}{t_{\frac{1}{2}}}$$

where $t_{\frac{1}{2}}$ is the half-time of "p" (or "o"). Once α, β, V_1, and V_2 are known, it is a simple matter to calculate F_1 and F_2 from Eqs. 8 and 9.

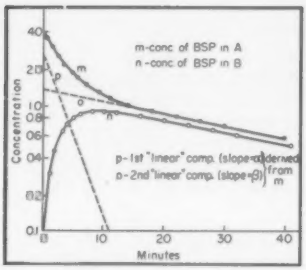

FIG. 3. Concentration-time curve derived from model after injection of BSP into compartment A (see expt. 3 in table)

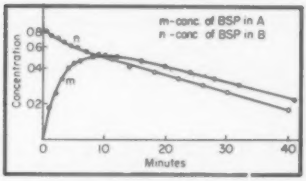

FIG. 4. Concentration-time curve derived from model after injection of BSP into compartment B (see expt. 4 in table)

A and B are vigorously mixed by forcing air into the bottom of each jar.

The labeling agent may be a dye or any other material which can be easily determined quantitatively, provided that it is chemically stable under the experimental conditions.* In our experiments we used bromsulfalein.

The model obeys the same mathematical relations as the ones derived for the illustration of phospholipide metabolism; i.e., V_1 = amount of water in tank A, y = amount of dye in tank A at any time, F_1 = rate of water flow from A to B and flow back from B to A, V_2 = amount of water in tank B, x = amount of dye in tank B at any time, and F_2 = rate of addition of water to B, which is equal to the rate of loss of fluid from V_2 into a sink.

In Fig. 3 the concentrations of bromsulfalein (BSP) in compartment A and B at various times after the introduction of the dye in A are shown as a semi-logarithmic plot. An examination of the curve "m" will show that it is the sum of two exponentials with coefficients α and β equal to the slopes of "p" and "o." The measurement of the slopes and calculation of rates F_1 and F_2 are outlined in Fig. 1.

* The color of many dyes depends upon the pH of the solution. To avoid the effect of pH changes due to aeration, it may be necessary to adjust the final pH of each sample before reading its optical density.

Comparison between Measured and Calculated Turnover Rates

Experiment No.	V_1 (ml)	V_2 (ml)	α $\times 10^2$	β $\times 10^2$	F_1 Measured (ml/sec)	F_1 Calculated (ml/sec)	F_2 Measured (ml/sec)	F_2 Calculated (ml/sec)
1	5080	11100	−4.69	−0.437	16.1	15.5	7.27	7.50
2	4930	11800	−4.92	−0.348	16.5	16.9	5.83	5.90
3	5470	10000	−5.52	−0.697	16.5	17.6	11.4	11.9
4	5320	25170	−4.10	−0.500	16.5	17.3	15.5	15.9
5	5310	25480	−4.06	−1.25	16.5	15.7	43.8	43.8
6	5600	25630	−3.69	−0.628	16.2	16.1	20.0	20.6
7	5220	26680	−1.68	−0.561	6.67	6.38	19.7	20.7

The table above shows both the measured and calculated "turnover rates" obtained from the model. The rates that were calculated from the dye disappearance curve agreed closely with those that were measured by collecting water at one of the outlets for a known time interval. This agreement indicates that the model behaves according to the assumptions used in the phospholipide problem.

In experiments 5, 6, and 7, we studied the effects of a change in the rates F_1 and F_2 on the concentration-time curves, all other factors remaining approximately constant. The volume V_2 was large compared to V_1, and the flow rates F_1 and F_2 were relatively small. Under these conditions, when F_1, the rate of exchange between compartment A and compartment B, was increased, the slope of the first linear component "p" (see Fig. 3) increased; the vertical distance between the concentration-time curves "m" and "n" decreased and the slope of "o" was little affected. On the other hand, when F_2 was increased, the slope of the first linear component "p" was little affected; the vertical distance between the concentration-time curves increased and the slope of the second linear component "o" was likewise increased.

It has been shown that when a compound has only one precursor, the specific activity of this compound and precursor are equal when the former reaches its maximum specific activity (*3*). Experiment 7 was performed to illustrate this principle. In this experiment, the label was introduced into compartment B. Thus we are dealing with a system where the dye in compartment A has only one immediate precursor, i.e., that in compartment B. Figure 4 shows the results. The two curves cross when the dye in compartment A has reached its maximum concentration.

A study of this type is of great help in interpreting specific activity-time curves derived from experiments on living organisms, when they are studied in different metabolic states.

The mathematical relations between turnover rates and the specific activity-time curves of the system become very complex as the number of compartments increases, as shown by the work of Sheppard and Householder (*4*). One can, however, easily build a three- or four-compartment model and study the concentration-time relations in each compartment after varying exchange rates or volumes one at a time. The information from a study of this type can be used to interpret specific activity-time curves derived from animal experiments.

An added advantage of the hydrodynamic model is that it provides a simple means of illustrating the isotope distribution in systems where a steady state is not maintained. This is achieved by adjusting the flow rates so that the volumes gradually change in the desired direction.

* * *

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Estimation of Neutron Energy for First Resonance from Thermal-Neutron Absorption Cross Sections

A quick estimate can be made, for reactor construction materials, of whether they have significant neutron absorption resonances. Results for 52 stable isotopes agree with the predictions of theory

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National Advisory Committee for Aeronautics, Cleveland, Ohio

INFORMATION CONCERNING neutron cross sections for stable isotopes in the rather wide neutron energy range occurring in nuclear reactors is sparse. This makes evaluation of the effect of reactor materials on the behavior of reactors quite difficult.

In particular, neutron absorption data are in many cases almost completely lacking; for most isotopes, only the thermal values and a few isolated higher-energy values are known. For many nuclei, neutron resonances occur at particular neutron energies, in the neighborhood of which very large cross sections are observed. As indicated by Harris *et al.* (*1*), appreciable portions of these resonances may be absorptive.

The theory of neutron capture would provide a means for computing cross sections if it were possible to carry out the necessary calculations with respect to the nuclear energy levels of the compound nucleus. Such calculations have proved, for the most part, to be impossible. The theory, however, does indicate that the absorption cross section below the first resonance varies inversely as the neutron velocity. Knowledge of the position and width of the first resonance would determine the neutron energy for which the inverse-velocity variation of absorption cross section ceases.

Examination of published data for some 52 stable isotopes indicates that it is possible to obtain an estimate of the neutron energy at which the first resonance occurs from the thermal-neutron absorption cross section. The empirical relation obtained from the experimental information is in qualitative agreement with the indications of a simplified version of the Breit-Wigner resonance theory of the nucleus.

Sources of Isotopic Data

The physical data for 34 isotopes, for which the thermal-absorption cross section, σ_{ath}, and the neutron energy for the first resonance, E_R, have been assigned with reasonable assurance, are presented in Table 1.

Neutron cross-section data were taken principally from Adair (*2*), in which the graphs of total cross section as a function of neutron energy served to locate generally the first few well-defined resonances. The specific energies at which the resonances occurred and the isotopic assignment were verified for most of the isotopes from the original papers referred to by Adair. A few elements listed, composed of a number of isotopes, contain a single preponderant isotope; the first strong resonance indicated on the total-cross-section curve was assigned to this isotope.

For elements for which fairly complete data are available, there is usually a small gap in the total-cross-section curve in the neutron energy range between the upper limit avail-

TABLE 1—Data on Thermal-Neutron Cross Sections and Resonance Energies

Isotope	Abundance (%)	σ_{ath} (barns)	E_R (ev)
He^4	100	0.006(4)*	1,100,000(2)*
Li^6	7.30	890†	270,000(2)
Be^9	100	0.0085(4)	625,000(2)
C^{13}	98.89	0.0035†	3,600,000(2)
N^{14}	99.64	1.86†	500,000(2)
O^{16}	99.76	0.001†	440,000(2, 3)
F^{19}	100	0.01(3)	35,000(2)
Na^{23}	100	0.47(4)	3,000(2)
Mg^{24}	78.98	0.027(7)	230,000(2)
Al^{27}	100	0.22(4)	2,300(3)
Si^{28}	92.19	0.08(7)	600,000(2)
S^{32}	95.06	0.48†	111,000(2)
Ca^{40}	96.92	0.0002(5)	255,000(2)
V^{51}	100	4.72(4)	2,700(1)
Mn^{55}	100	12.8(4)	345(2)
Co^{59}	100	34.2(4)	120(2)
Zn^{67}	4.12	1†	480(1, 2)
Ge^{73}	7.9	13.7(4)	95(1)
Rh^{103}	100	150(4)	1.30(2, 3)
Pd^{108}	26.7	11(5)	24(3)
Ag^{107}	51.35	30.0(6)	16(2, 3)
Ag^{109}	48.65	83.7(6)	5.1(2, 3)
Cd^{113}	12.34	24,000(5)	0.17(2, 3)
Cd^{116}	7.66	1.40(5)	110(3)
I^{127}	100	6.06(4)	20.3(2, 3)
Sm^{149}	13.84	65,000(3)	0.096(2, 3)
Sm^{152}	26.63	135(3)	10.0(2, 3)
Eu^{153}	52.23	240(3)	0.465(3)
Gd^{157}	15.71	240,000(3)	0.031(2, 3)
Ta^{181}	100	21.3(4)	4.1(2, 3)
W^{186}	28.64	34(5)	18(2, 3)
Ir^{191}	38.5	1,000(5)	0.620(2, 3)
Ir^{193}	61.5	130(5)	1.29(2, 3)
Au^{197}	100	95(4)	4.8(2, 3)

* Numbers in parentheses refer to sources of the data.

† Estimated from σ_{ath} values for other isotopes of the element and from relative isotopic abundances.

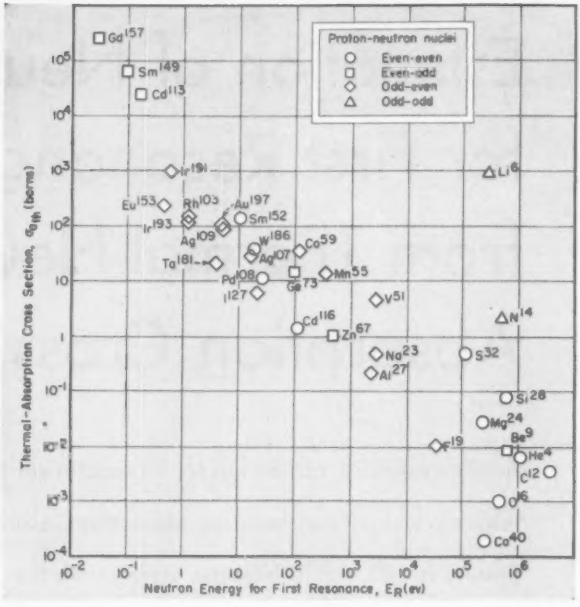

FIG. 1. Thermal-neutron absorption cross section as a function of neutron energy for first resonance

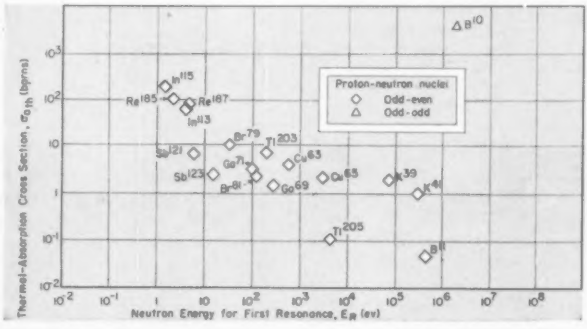

FIG. 2. Thermal-neutron absorption cross section as a function of neutron energy for first resonance for paired isotopes

able from slow-neutron spectrometers and the lower limit of particle-accelerator-neutron sources. Where values of total cross sections at these limits matched, it was assumed that no resonances occurred in this gap.

Isotopic thermal-absorption cross sections and relative isotopic abundances of the elements were taken from five sources (*3–7*); the pile-oscillator values of thermal-absorption cross section were chosen wherever available. A few unknown isotopic thermal-absorption cross sections were estimated from the known thermal-absorption cross sections for the other isotopes of the element and the relative isotopic abundances. Additional resonance data were obtained from other sources (*8–13*).

Data on nine elements, each consisting principally of two isotopes with known thermal-absorption cross sections and having well-defined resonance peaks on the total-cross-section curves, are presented in Table 2. The lowest two resonance peaks have been assigned to the two individual isotopes in accordance with an empirical relation to be observed from the data of Table 1.

Correlation of Data

The dependence of the thermal-absorption cross-section, σ_{ath}, (corresponding to an energy of 0.025 ev) on the neutron energy for the first resonance, E_R, for the isotopes of Table 1 is presented in Fig. 1. The even-even,

even-odd, odd-even, and odd-odd nuclei are indicated as such.

With the exception of the two odd-odd lithium and nitrogen nuclei, there is a general correlation between σ_{ath} and E_R. It is apparent that E_R increases with decreasing σ_{ath} rather irregularly. It is, however, impossible to draw any conclusions concerning differences in the behavior of even-even, even-odd, and odd-even nuclei.

The dependence of σ_{ath} on E_R for the nine pairs of isotopes of Table 2 is presented in Fig. 2. The well-defined resonances for each element have been assigned to the isotopes of the element in accordance with the general relation represented in Fig. 1. All of these isotopes happen to be odd-even nuclei with the exception of the odd-odd boron isotope. Again, although the scatter in the data is large, the same general relation holds.

Analysis by Inglis (*13*) of the B^{10} neutron-cross-section data indicates the presence of resonances at energies below the well-defined resonance at 2 Mev; Inglis suggests that a number of resonances combine to form a broad peak near 100,000 ev. For energies in the region of 100,000 ev, the odd-odd boron isotope still departs significantly from the other nuclei.

A reason for the large scatter at the high-energy resonances may be the following: The low-energy first resonances are observed principally in heavy nuclei for which neutron capture with gamma or beta emission is the dominant absorption reaction. The neutrons involved have minimum angular momentum, and resonance processes involving them would be expected to be related to the thermal cross section, which is also due to these *s*-neutrons. The high-energy first resonances occur mainly in light nuclei and involve neutron capture with emission of the various particles. The neutrons involved may possess one or more units of angular momentum, so the resonance processes are not expected to be related to the thermal-absorption process. For example, as cited by Peshkin and Siegert (*14*), the resonance absorption of neutrons by Li^6 with alpha emission at energies of about 250,000 ev is known to be due to *p*-neutrons (neutrons with one orbital unit of angular momentum).

The data of Figs. 1 and 2 are replotted in Fig. 3 for comparison. The paired-isotopic data fall within the scatter of the data of Fig. 1. Except for the three odd-odd nuclei Li^6, B^{10}, and N^{14}, all of the available isotopic data have some measure of correlation.

Additional contributory evidence of the validity of the general relation indicated in Fig. 3 is presented in Table 3. The resonance energies qualitatively indicated in the table fall within the scatter of the data in Fig. 3.

Neutron Resonance Theory

The general dependence of σ_{ath} on E_R shown in Fig. 3 may be explained by the theory of excited states of the compound nucleus. Development and application of this theory are given by Bethe (*15*) and by Feshbach, Peaslee, and Weisskopf (*16*). It will be shown that after introducing some fairly general experimental information into the theory, the theory approximates the experimental data concerning the individual nuclei.

The cross section for an absorption or scattering process as a function of the energy of the incident particle

TABLE 2—Data on Paired Isotopes

Isotope	*Abundance (%)*	σ_{ath} *(barns)*	E_R *(ev)*
B^{10}	18.83	3,800*	2,000,000(*2*)†
B^{11}	81.17	0.05(*5*)†	430,000(*2*)
K^{39}	93.08	2.13*	70,000(*2*)
K^{41}	6.91	1.0(*5*)	305,000(*2*)
Cu^{63}	69.09	4.3(*3*)	570(*3*)
Cu^{65}	30.91	2.1(*3*)	3,000(*3*)
Ga^{69}	60.2	1.5(*3*)	278(*8*)
Ga^{71}	39.8	3.4(*3*)	100(*8*)
Br^{79}	50.57	10.9(*5*)	34(§)
Br^{81}	49.43	2.25(*5*)	125(§)
In^{113}	4.16	58(*5*)	3.8(*2, 3*)
In^{115}	95.84	197(*5*)	1.44(*2, 3*)
Sb^{121}	57.25	6.8(*5*)	5.8(*2, 3*)
Sb^{123}	42.75	2.5(*5*)	15(*2, 3*)
Re^{185}	37.07	101(*5*)	2.15(*2, 9*)
Re^{187}	62.93	75(*5*)	4.5(*2, 9*)
Tl^{203}	29.52	7.6(*5*)	230§
Tl^{205}	70.48	0.11(*5*)	4,300§

* Estimated from σ_{ath} values for other isotopes of the element and from relative isotopic abundances.

† Numbers in parentheses refer to sources of the data.

§ Data obtained at Columbia University.

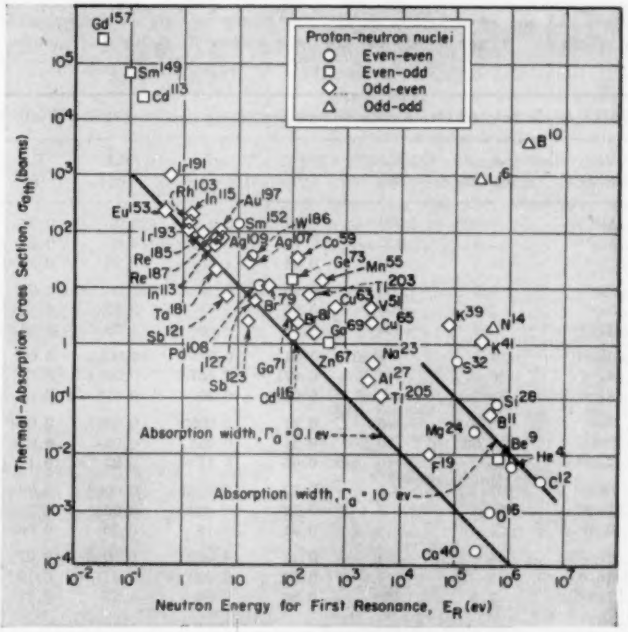

FIG. 3. Thermal-neutron absorption cross section as a function of neutron energy for first resonance. Straight lines are plots of Breit-Wigner single-level formula

TABLE 3—Additional Evidence for the Relationship in Fig. 3

Isotope	*Abundance* (%)	σ_{ath} (*barns*)	*Resonance energy*
$_{18}A^{40}$	99.60	0.60(3)*	Resonance above 10,000 ev suggested(*10*)
$_{21}Sc^{45}$	100	11.8(4)	Resonance cited at 1,000–10,000 ev(*1*)
$_{33}As^{75}$	100	4.14(4)	Resonance cited at 100–1,000 ev(*1*)
$_{40}Zr^{90}$	51.46	0.11(3)	Resonance dip at 800,000 ev may be due to most abundant isotope(*11*)
$_{41}Nb^{93}$	100	1.06(4)	No resonance apparent below 1,000 ev(*12*)
$_{57}La^{139}$	100	8.8(4)	No resonance apparent below 700 ev(*2*)
$_{83}Bi^{209}$	100	0.015(3)	No resonance apparent below 1.4 Mev(*2*)

* Numbers in parentheses refer to sources of the data.

near an isolated resonance is given by the Breit-Wigner single-level formula. Inasmuch as the data in question are concerned with energies below the first level, the single-level formula will essentially give correct results down to very low energies.

The absorption cross section, σ_a, for a neutron of kinetic energy E incident upon a nucleus with a resonance level occurring at a neutron energy E_R (both energies measured in the center-of-mass system) is given by

$$\sigma_a = \pi\lambda\lambda_R g \left[\frac{\Gamma_a \Gamma_n}{\Gamma^2/4 + (E - E_R)^2}\right] \quad (1)$$

where $2\pi\lambda$ and $2\pi\lambda_R$ are the neutron wavelengths corresponding to the energies E and E_R, respectively, and Γ_a and Γ_n are the energy widths for the emission of a γ-ray or a charged particle (neutron capture) and the emission of a neutron (scattering), respectively, at the energy E_R. The total width, Γ, is the sum of the partial widths, Γ_a and Γ_n, which are measures of the relative probability of neutron absorption and scattering.

The quantity g is a statistical-weight factor determining the fraction of the total resonance states of the compound nucleus corresponding to a particular process. If a neutron has no angular momentum with respect to the nucleus by which it is captured, which is usually the case, and if this nucleus has the spin I, then

$$g = \tfrac{1}{2}[1 \pm 1/(2I + 1)] \quad (2)$$

For large I, the factor g has a value of about $\frac{1}{2}$; for $I = 0$ only the plus sign is significant and g has its maximum value of 1; it has its minimum value of $\frac{1}{4}$ for $I = \frac{1}{2}$. If, for simplicity, a value of $g = \frac{1}{2}$ is introduced in Eq. 1, the largest error resulting in σ_a is a factor of 2.

The remaining quantities affecting σ_a are the neutron-absorption and scattering widths, Γ_a and Γ_n. Empirical and theoretical rules for estimating these values for various nuclei are presented by Bethe (*15*). The absorption width, Γ_a, corresponding to the emission of γ-rays or of charged particles has been inferred from experimental data. Bethe (*15*) has summarized the results for widths corresponding to γ-ray emission. His results indicate that the Γ_a are of the order of 0.1–1 ev for most of the medium-heavy nuclei; for light nuclei (as obtained from proton-capture data), Γ_a is of the order of 1–10 ev. An approximate expression for the neutron width may be obtained from experimental data summarized by Bethe (*15*) and Wigner (*17*) and from the theoretical discussion by Feshbach *et al.* (*16*) and Weisskopf (*18*). The results can be represented by

$$\Gamma_n = C\sqrt{E_R}\, D \quad (3)$$

where C is constant for a given nucleus and varies from 10^{-4} to 10^{-3} $(ev)^{-\frac{1}{2}}$ for various nuclei, and D is the resonance-level spacing.

The resonance-level spacing, D, is not known for most nuclei. If, however, D is taken as being approximately equal to the kinetic energy, E_R, of the neutrons corresponding to the first resonance, Eq. 3 becomes

$$\Gamma_n = C\, E_R^{3/2} \quad (4)$$

To check the values of Γ_a and Γ_n given, and to determine the C of Eq. 4 more precisely, use is made of the experimental data of Harris *et al.* (*1*) wherein measurements of resonance scattering and resonance absorption integrals, and average values of the resonance scattering fraction, Γ_n/Γ, are presented for a number of elements and isotopes. From these data, values of $g\Gamma$ have been computed; by assuming a value of $g = \frac{1}{2}$ (for reasons explained previously) the width values Γ_n and Γ_a have been calculated for

TABLE 4—Calculated Neutron Resonance-Scattering and Absorption Widths*

Isotope	σ_{ath} (*barns*)	E_R (*ev*)	$\frac{\Gamma_n}{\Gamma}$	$g\Gamma$ (*ev*)	Γ_n† (*ev*)	Γ_a† (*ev*)
$_{13}Al^{27}$	0.22	2,300	0.99	11.8	23.3	0.24
$_{25}Mn^{55}$	12.8	345	0.99	12.5	24.8	0.25
$_{27}Co^{59}$	34.2	120	0.94	1.58	2.96	0.19
$_{29}Cu^{nat}$	3.6	~1,000	0.95	9.40	17.9	0.94
$_{31}Ga^{nat}$	2.8	~200	0.95	1.35	2.57	0.13
$_{33}As^{75}$	4.1	~500	0.72	0.89	1.28	0.50
$_{45}Rh^{103}$	150	1.30	0.043	0.0053	0.0005	0.01
$_{47}Ag^{107}$	30.0	15	0.071	0.067	0.0095	0.12
$_{47}Ag^{109}$	83.7	5.1	0.038	0.20	0.015	0.39
$_{51}Sb^{nat}$	5.3	~5	0.21	0.0050	0.0021	0.008
$_{53}I^{127}$	6.06	20.3	0.31	0.055	0.034	0.076
$_{62}Sm^{152}$	135	10	0.66	0.17	0.22	0.11
$_{72}Hf^{nat}$	102	~1	0.17	0.0023	0.0008	0.004
$_{73}Ta^{181}$	21.3	4.1	0.12	0.020	0.0047	0.034
$_{74}W^{186}$	34	18	0.81	0.16	0.26	0.061
$_{75}Re^{nat}$	84	~3	0.11	0.012	0.0026	0.021
$_{79}Au^{197}$	95	4.8	0.14	0.060	0.017	0.10
$_{81}Tl^{203}$	7.6	230	0.80	0.80	1.27	0.32

* Calculated from experimental data of Harris *et al.* (*1*).
† Calculated using statistical-weight factor of $g = \frac{1}{2}$.

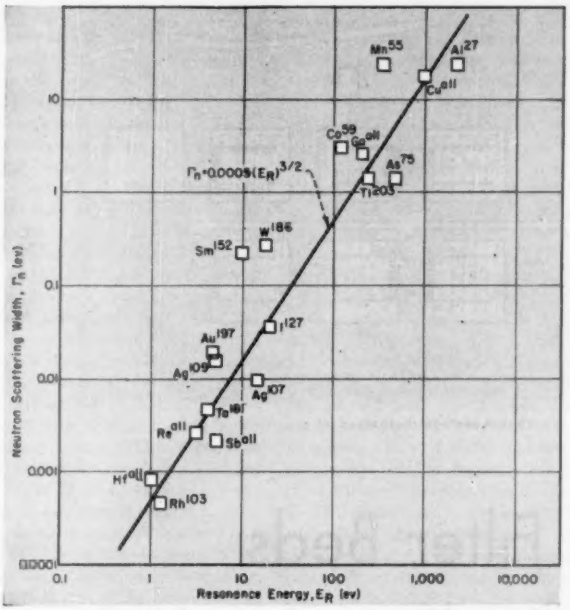

FIG. 4. Calculated variation of neutron scattering width with resonance energy for value of $g = \frac{1}{2}$

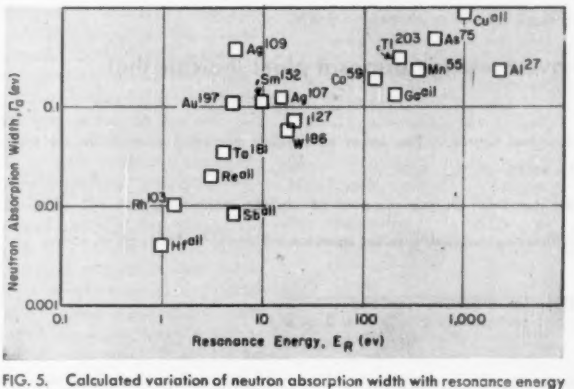

FIG. 5. Calculated variation of neutron absorption width with resonance energy for value of $g = \frac{1}{2}$

these elements and isotopes. These data are presented in Table 4. The widths Γ_n and Γ_α are also represented as functions of E_R in Figs. 4 and 5, respectively. The dependence of Γ_n on E_R is reasonably well represented by Eq. 4 with $C = 5 \times 10^{-4}$ $(\text{ev})^{-\frac{1}{2}}$ which is represented by the straight line in Fig. 4. The absorption widths, Γ_α, increase with E_R as indicated in Fig. 5; the average is about 0.1 ev as claimed by Bethe.

If the absorption cross section is measured in barns, and neutron energies and widths are measured in electron volts, the single resonance-level formula of Eq. 1 may be written

$$\sigma_a = \frac{65.8 \times 10^4\, g}{\sqrt{E}\sqrt{E_R}} \left[\frac{\Gamma_a \Gamma_n}{\Gamma^2/4 + (E - E_R)^2} \right] \tag{5}$$

Using a value of $g = \frac{1}{2}$ and width Γ_n as given by Eq. 4 with a value of $C = 5 \times 10^{-4}$, the absorption cross section, σ_a, is given by

$$\sigma_a = \frac{165\, \Gamma_a E_R}{\sqrt{E}\,[(\frac{1}{4})(5 \times 10^{-4} E_R^{3/2} + \Gamma_a^2) + (E - E_R)^2]} \tag{6}$$

For resonance energies above 1 ev, the effect of the total width in Eq. 5 or 6 is negligible, so that the thermal-absorption cross section, σ_{ath}, evaluated at $E = 0.025$ ev may be given by the Breit-Wigner single-level formula

$$\sigma_{ath} = 1{,}040\, \Gamma_a / E_R \tag{7}$$

Inasmuch as Γ_a is almost independent of E_R, Eq. 7 provides the reason for the correlation between σ_{ath} and E_R.

Equation 7, with the assumption of $\Gamma_a = 0.1$ ev, is plotted in Fig. 3 and appears to agree fairly well with the experimental resonance data of the medium and heavy nuclei where the assumptions made are most valid. Equation 7, with $\Gamma_a = 10$ ev, as appropriate for the case of light nuclei —in accordance with Bethe's summary of experimental data (*15*)—is also plotted in Fig. 3 and is in fairly good agreement with the experimental results obtained for light nuclei.

The isotopes Gd^{157}, Sm^{149}, and Cd^{113} have resonance energies which are small compared with their widths, so that Eq. 1 does not apply. The thermal-absorption cross section at 0.025 ev for Sm^{149} is an average over the thermal spectrum of neutrons and so includes considerable contribution from the resonance peak. It is not surprising, therefore, that these isotopes (Fig. 3) show cross sections in excess of that given by Eq. 7.

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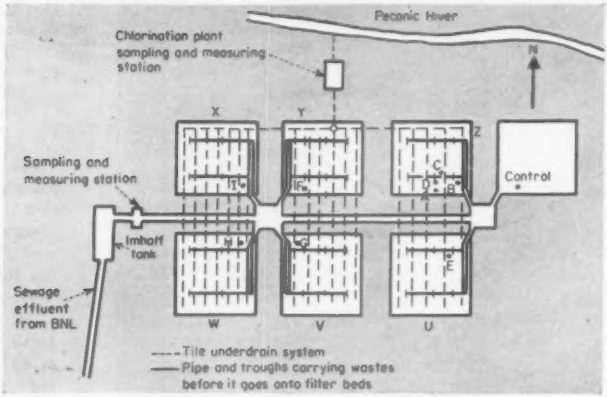

LAYOUT of the Brookhaven sewage-disposal plant

Efficiency of Filter Beds for Treating Radioactive Waste

Studies conducted at the Brookhaven sewage disposal plant indicate that:

- **Pure sand removes very little activity from waste. The layer of foreign material laid down by the sewage is responsible for most of the removal**
- **The bacterial content of this layer is important in the removal of certain radioisotopes**
- **Apparent fluctuations in filter-bed efficiency probably occur because of washing-through of radioactivity in the bed**
- **The efficiency of the beds varies widely for different isotopes**

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CONSIDERABLE INTEREST attaches to the performance of conventional sewage treatment plants in removing radioactive contaminants present in small amounts in wastes resulting from research, industrial or medical uses of isotopes.

The disposal plant at Brookhaven National Laboratory affords an excellent opportunity for engineering studies of the adsorptive properties of sand filter beds. Although the conditions at this plant may not be typical, since the sewage here has an unusually low concentration of suspended solids, the results obtained here might be of interest as a general guide to what may be expected.

BNL Disposal System

At Brookhaven, the sanitary sewage, the ordinary laboratory wastes and the low-level radioactive wastes from local hold-up systems, all flow together through a common tile pipe to the waste disposal area located about two miles northeast of the pile complex.

The disposal plant was built by the military during World War II, and was again put into operation by the laboratory staff when the site was taken over early in 1947. It consists primarily of an Imhoff tank for sludge settling and a system of six intermittent sand filters of about one acre each. Although the volume of waste varies from day to day, a 12-month average shows a flow of about 223,000 gallons per day.

The diagram shows the general layout of the BNL sewage disposal plant. The liquid wastes are sampled after leaving the Imhoff tank by a trebler-type proportional sampler. The volume, in gallons per day, is measured by a recording meter. These data make it possible to calculate the specific activity of the liquid waste in curies/cm³ before it goes onto the filter beds.

The liquid is automatically dumped onto one of the filter beds from a dosing tank, after reaching the tank capacity of 25,000 gallons. A fairly even distribution is reached over the entire bed by a system of open troughs.

In the filter bed, the liquid soaks down through 6 ft of sand and gravel, and most of it is picked up by a tile underdrain system. The water continues to flow by gravity to a stilling tank where it is again sampled and measured, and then chlorinated.

The waste is finally discharged to a small stream, which is usually dry during July, August, and September. This stream, one of the headwaters of the Peconic River, reaches the Peconic Bay as a fairly sizable river at Riverhead, N. Y.

If the amount of activity that goes onto the filter beds, and the amount that remains in the liquid picked up by the underdrains are known, a fairly good estimate can be made of the amount that is lost to the soil and ground water. During seasons of low rainfall, as much as 30% of the liquid is lost, while during periods of considerable rain, the tile field picks up nearly as much liquid as goes onto the filter bed.

The purpose of these studies was to determine the efficiency of these filter beds in removing some of the most common radioisotopes from the laboratory low-level sewage. It should be said at the outset that the behavior of a 6-ft bed of sand, with adhesive organic growths and clay particles as a decontaminating facility, is difficult to evaluate. The data obtained must be considered incomplete, but certain significant conclusions are possible. The results have been interpreted only for the local situation at Brookhaven, and they do not necessarily apply to similar situations at other sites.

Filter-Bed Spiking

The first series of tests consisted of actually "spiking" one of the filter beds. The *Z* bed (diagram, p. 44) was chosen for all tests in order to eliminate such differences as bed composition and drainage conditions. The sewage was essentially cleaned, as far as radioactivity was concerned, by holding all laboratory wastes in hold-up tanks for several days before and after the spiking. In that way, practically all the activity that went onto the bed was due to a known quantity of a specific isotope which was applied during the dumping of a single 25,000-gallon dosing tank.

The radioisotopes applied in spiking were: P^{32}, in the form of HP_3O_4; I^{131}, as an iodide in weak basic sodium sulfite solution; Sr^{90}, as acid chloride; and six-month-old mixed fission products, as nitrates.

Approximately 200,000 gallons of sewage per day flowed onto this bed in eight equal dosings. In addition to the proportional sample covering each day's operation, samples were taken at regular intervals of the water picked up by the tile underdrain system. Each sample was assayed for activity. If all these activities are integrated, the amount of activity coming through the filter bed can be calculated. The difference between the activity that is put on the bed and that which comes through remains either on the sand or is lost in the ground water not picked up by the tile underdrain system. Table 1 shows the results of these tests.

Several interesting facts were noted during these tests: In the case of P^{32}, the activity concentration coming through the sand reached its greatest value 24 hr after the spike, and then "drooled off" for the next 5 days. With the other isotopes and canal water, however, the highest concentration was almost immediately following the spike, and fell off rapidly, reaching background within 3 days. The composition of the pile canal water is a mixture of many isotopes. Its main component, which is anionic, is adsorbed much less readily than the cations.

Laboratory study of filter beds. Since the radioactive ions used in these

TABLE 1—Results of Spiked Tests on Filter Bed *Z*

Element	*Amount applied (mc)*	*Amount picked up by tile underdrain system (mc)*	*Lost to the sand and ground water (mc)*	*Percent removal of activity*
P^{32}	10	0.8	9.2	92
I^{131}	11.6	1.4	10.2	88
Mixed fission products	11.6	0.2	11.4	98
Sr^{90}	10.2	0.1	10.1	99
Pile canal water	52.4	27.7	24.7	47

TABLE 2—Activity Adsorption by Filter-Bed Layers at Various Points in the Disposal System

Location	*Top 3 in.* (dpm)*	*Percent of total*	*Next 9 in. (dpm)*	*Percent of total*	*Below 1 ft (dpm)*	*Percent of total*
A	369	88	42	10	11	2
B	5,190	88	530	9	175	3
C	2,630	93	136	5	38	2
D	4,860	87	500	9	206	4
E	192	92	12	6	5	2
F	93	85	9	8	7	7
G	94	88	8	7	5	5
H	117	100	0		0	
I	86	100	0		0	
Control	135	100	0		0	

* Control has been subtracted from this column. Control varies with time and concentration of airborne radioactive particles following atomic bomb tests

tests tend to adsorb onto particles of clay, organic compounds, etc., mixed with the filter-bed sand, a study was conducted in the laboratory to determine the location of this activity.

Various test holes were dug in the filter beds. The top 3 in. of the filter bed was much darker than the rest of the sand, so it undoubtedly contained most of the organic and inorganic compounds laid down by the sewage. The next 9 in. showed some slight darkening. The remaining 5 ft of sand and gravel down to the tile underdrains seemed to be essentially clean. Therefore, the top 3 in., the next 9 in. and the remaining lower level were arbitrarily chosen as the three layers to investigate.

Columns measuring 5.6 cm in diameter were filled to a depth of 109 cm with one of the three sand layers of filter bed taken from different locations. Since preliminary tests showed that 3 *N* nitric acid removed over 95% of the radioactivity adsorbed on the sand, each column was treated with 500 ml of this acid. More acid brought down no more activity and tended only to dilute the effluent.

The acid effluent was then counted in a jacketed tube. Samples were also processed and counted in a gas-flow counter. The results of counting in the flow counter were consistently 30% higher for all samples than were found using the jacketed tube. This increase is presumably due to soft beta components that would be screened out by the glass of the jacketed tube. However, the relative values for various positions in the bed were about the same for both methods of counting. Table 2 gives the results of the tests using the jacketed tube data. Locations can be spotted by referring to the diagram on p. 44.

The *Z* filter bed was used for all the spiked test runs. The readings for locations *A*, *B*, *C* and *D* were much higher than for other points because 45 mc more of long-lived radioactivity is adsorbed to bed *Z* than to any other.

These results show quite conclusively that approximately 90% of the total activity adsorbed in the filter beds remains in the top 3 in. The next 9 in. holds about 8%, and the remaining 5 ft contains only 2%.

Laboratory Saturation Studies

The filter beds, or parts of them, might become saturated and refuse to hold up more of certain isotopes. To investigate this problem, a number of columns were set up in the laboratory duplicating, as closely as possible, the actual make-up of the filter bed. A large volume of sewage was spiked with mixed fission products over six months old. A column was dosed every 6 hr with 300 ml of this sewage containing 0.14 μc of activity. The volume and time interval duplicate the dosing conditions that occur at the filter beds.

The data showed that for the first 16 dosings, covering about 4 days, the column removed approximately 98% of the activity. During the next 10 dosings, covering 2½ days, the efficiency dropped to about 70% removal. It was noted that the length of time required for the sewage to flow through the column gradually increased. On the 37th dosing, or at the end of 9 days, about 1½ hr were required for the liquid to pass through the column—as compared to 5 min for the first dose—and only 20% of the activity was removed.

After 24 hr, the unused sewage showed a brown flocculent precipitate. Even though the mixture was shaken and mixed before each dosing, the presence of part of the activity in the precipitate indicated that this test was probably not representative of actual operating conditions. To overcome this precipitation, the tests were repeated using fresh sewage brought in daily, and equal amounts of it were spiked with I^{131}, P^{32} and 6-months-old mixed fission products. All assays took into account the radioactive decay of I^{131} and P^{32}.

In the case of P^{32}, the column never removed less than 85% of the activity during the first 7 days. During this period, it had been dosed 28 times with a total of 400 μc. Even though the doses during the next 4 days were much smaller, the efficiency of the column dropped to 60%. This drop in efficiency was probably due, at least in part, to the washing down of activity which was loosely adsorbed in the bed.

On the 12th and 13th days, 1.3 mc of activity was put on the column, and the efficiency rose to 94% removal. This increase in removal efficiency, as a result of the large dose, can probably be explained as the inverse of the reduction in efficiency noted above when the dosing rate was reduced. That is, if some washing-through occurs for repeated applications, such washing-through will be smaller for a single large dose than for the equilibrium situation after repeated large doses. There may also be small biological changes in the column contributing to greater or lesser hold-up. It appears, therefore, that the column is capable of handling considerably larger amounts than the 2.2 mc of P^{32} applied before reaching the saturation point.

A second column received 2 mc of I^{131}. Until the 37th dose, or the 9th day, the rate of activity removal was almost constant—the efficiency did not fall below 80%. After the 37th dose, the column started to lose its retentive ability. However, its removal efficiency never fell below 50%, even though 1 mc was added on the 13th day.

The mixed-fission-products column was dosed with 6 mc of activity over a period of 10 days. There were fluctuations between dosings; however, efficiency was high for the first 5 days and fell gradually. It varied from 99% removal on the 5th day to 40% on the 10th day. This up-and-down fluctuation in efficiency is also noted in the daily log of the performance of the filter beds at the sewage plant.

On the basis of these accelerated tests, it may be concluded that, under the present low-level dosing conditions (not exceeding an average of 3×10^{-12} curies/cm^2), it is unlikely that the filter beds would become saturated to the point that the activity-removal efficiency would drop below 50%. A dose of 2 mc on one column, which is 6.2×10^{-7} acres in area, corresponds to 3,200 curies for the entire area of each bed. Thus, neglecting decay, it would take over 1,000 years, at the present rate of dosing, to reach even a semblance of saturation.

The foregoing analysis leaves much to be desired because of uncontrolled and unknown variables in this complex chemical phenomenon.

* * *

This article is based on a paper presented before the Radiation Division, American Industrial Hygiene Association, Cincinnati, Ohio, April 24, 1952. The author wishes to acknowledge the work done on these projects by Mr. Thomas Burns, Rensselaer Polytechnic Institute, summer appointee to BNL staff, 1951, and by Mr. Bryant Hanson, Chemical Processing Plant, American Cyanamid Co., Arco, Idaho. Research was carried out under the auspices of the U. S. Atomic Energy Commission.

Film Measurement of Beta Radiation Dose

The question of the validity of using film-density values to determine beta radiation exposure often arises where various absorption conditions exist. In work with uranium, it has been shown that the film method is reliable

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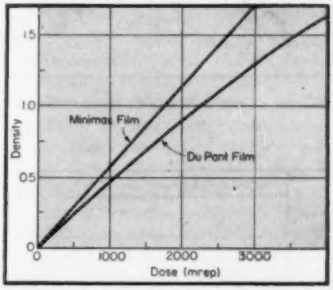

FIG. 1. Typical characteristic density-dose curves for films exposed to beta radiation from uranium

IN PERSONNEL MONITORING where the film method of measuring beta radiation dose is used, it is important to know the possible variations of film response with the energy of the beta particles. To check on the monitoring system in effect at this laboratory, we investigated these variations and found that our calibration system was adequate.

Beta Dose Measurement

Beta dose is defined in terms of energy loss by ionization in air, and fundamental dose measurements must be made with air ionization chambers. To use film for this purpose, it is necessary first to establish a relationship between fundamental dose measurements and the film blackening produced by such exposures. This calibration can then be used to convert observed film blackening, measured as optical density, to dose.

The required density-dose relationship is constructed as follows:

1. The dose rate at the surface of a source is determined by an air ionization measurement.

2. Films are exposed to the standardized source for different lengths of time.

3. The observed film densities are plotted against the corresponding doses to form a characteristic density-dose curve.

In connection with item 1 in the procedure, our laboratory uses an extrapolation chamber for the air-ionization measurement. As the standard source, we use uranium because our work is concerned, in the field, with exposure to the beta particles emitted primarily by the uranium daughter Pa^{234} (UX_2), 2.3-Mev maximum energy.

Two examples of calibration curves (item 3) are shown in Fig. 1 for uranium. Densities of films worn in the field are converted to dose using these curves. The film dose measurement is therefore based on a standardization with a primary instrument for one particular source.

Affecting Conditions

Under actual work conditions, the UX_2 beta radiation undergoes various degrees of absorption, so that the effective sources differ from the standardizing source. As a result, this calibration may not be valid. The variation of ionization with beta-particle energy may be different in air from that in the emulsion. For X-rays, film response is known to be strongly dependent upon wavelength. If film is also energy-dependent for beta radiation, equal doses (as measured by air ionization) from beta sources of different energies will produce different film densities.

The energy distribution of the

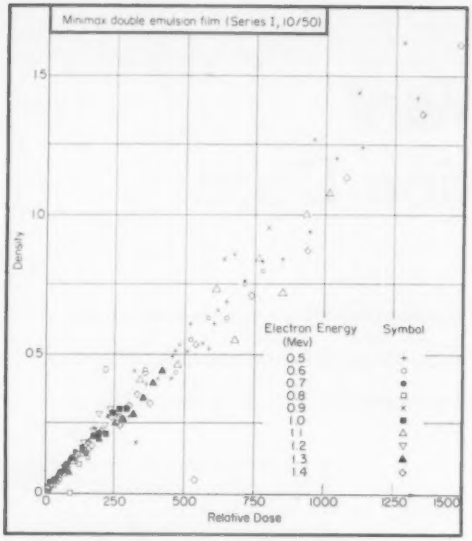

FIG. 2a. Density-dose plot, Minimax film (see Fig. 2b)

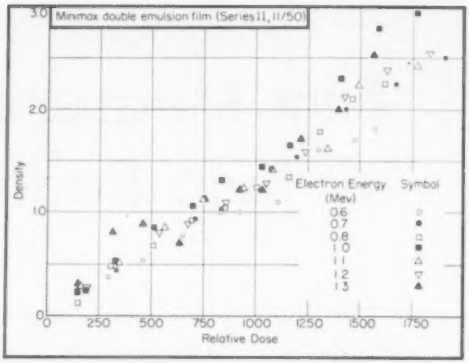

FIG. 2b. Density-dose plot, second series of Minimax film (see also Figs. 2a and 2c). All three graphs on this page are exposures to monoenergetic electrons

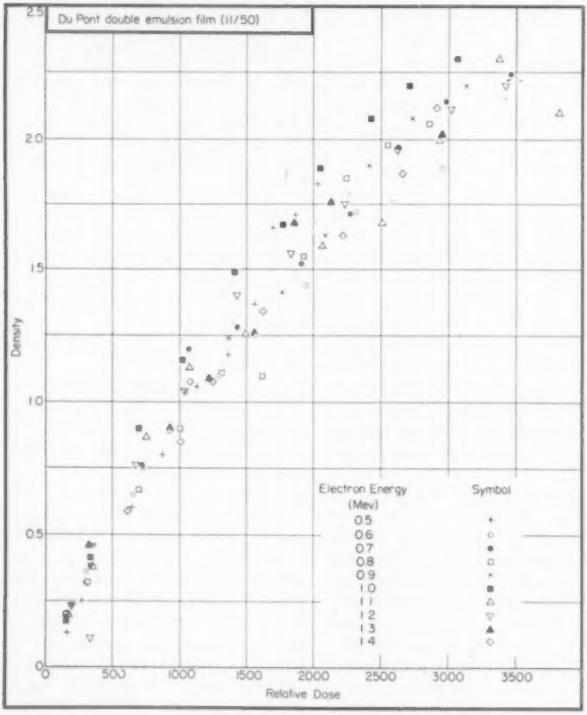

FIG. 2c. Density-dose plot, Du Pont film (see others in Figs. 2a and 2b)

absorbed UX_2 radiation exposure under field conditions may differ from the spectrum at the surface of uranium metal. If the film is energy-dependent, it will respond differently to this altered distribution.

If these effects are significant, the density-dose relationship determined for the unabsorbed radiation will not hold. To interpret film density as dose under such conditions, it is necessary to know the energy distribution of the ionizing radiation and establish calibrations for the different distributions.

An additional consideration is that a film badge is ordinarily worn for a work week and represents an integrated exposure to the many different absorption conditions encountered. In such an instance, the calibration of film for the field measurement of beta-radiation dose would be impractical.

To determine the validity of the present film method, then, it is necessary to investigate the significance of the possible sources of error.

Experimental

In studying the possible variation of film response with energy and absorption, two methods of investigation were employed. In the first, using monoenergetic electron beams, the variation of film density with beta dose from particles of different energy was studied. In the second, the beta radiation emitted from a uranium metal source was absorbed through different filter thicknesses and the film

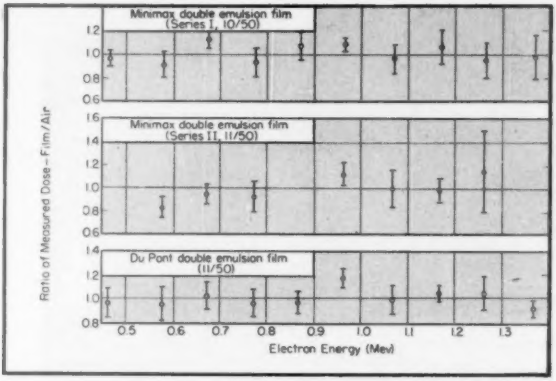
Minimax double emulsion film (Series I, 10/50)
Minimax double emulsion film (Series II, 11/50)
Du Pont double emulsion film (11/50)
Ratio of Measured Dose—Film/Air
Electron Energy (Mev)

FIG. 3. Plot showing standard deviation of the ratios of film to air dose-measurements for films exposed to monoenergetic electrons

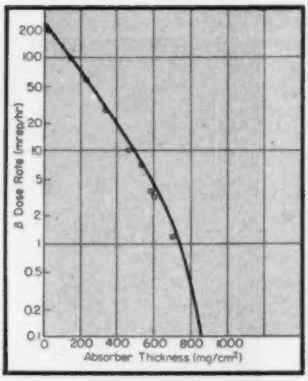

FIG. 4. Effect of absorption on film dose-measurements (UX_2 radiation, Al filters)

and air ionization measurements for the different source conditions compared.

Monoenergetic Electron Beam

At the National Bureau of Standards, Washington, D. C., film packs were exposed, in the badges in which they are worn, to monoenergetic beams directed normal to the film face. The energies of the beams used covered the range of the accelerator, 0.5–1.4 Mev. The relative dose at the exposure position was measured with an extrapolation chamber.

At each energy, a series of films from the same emulsion batch was exposed to the beam for different lengths of time, then developed and read at this laboratory. Du Pont type 552 and Minimax double emulsion films were used. In Fig. 2, the densities are plotted against measured dose for each energy series.

The dose was estimated, using standard calibration curves such as those described earlier. The dose thus obtained was compared with the observed dose, measured by air ionization. For each energy series, the ratios of film to air dose measurement were averaged. These average ratios are shown in Fig. 3, plotted against the actual energy of the electrons incident upon the film. The indicated ranges are the standard deviation of the ratios in each group. These results are for the same data as shown in Fig. 2.

From the comparison of dose ratios for the different energies, there is no apparent deviation due to the beta-particle energy between film and standard air measurements of dose in the 0.5–1.4 Mev range. From this result, it may be concluded that, for this range, film ionization is proportional to ionization in air.

Absorbed Beta Radiation

The standard calibration curve discussed previously was constructed by exposing film for different lengths of time to the surface of a uranium source. The dose rate for this condition was determined and the dose therefore known. The same curve, however, is used to interpret, as dose, film densities that are known to result from exposure to the softened radiation encountered under field conditions. To determine whether the film calibration is valid under these conditions, air-ionization and film dose measurements were compared for UX_2 radiation which had passed through absorbers.

Aluminum absorbers were used to approximate the air absorption in the field. Using the extrapolation chamber in this laboratory the beta dose rate from uranium through various thicknesses of aluminum was measured. Films were then exposed, for different times, through the same aluminum filters, and the densities converted to dose using the calibration curve. Figure 4 shows the relationship between the dose rates determined by the two measurements. The experimental points represent several different sets of film experiments and many replicate determinations.

For a large part of the range studied, the agreement is good. At the conditions of greatest absorption there is some tendency for the film measurement to be low. The importance of this apparent deviation depends upon how much of the weekly dose is received with high absorption.

The worst error is incurred at the lowest dose rates but, at such levels, the difference is not of practical concern. For example, assuming a condition where all the exposure is to greatly absorbed radiation at a distance in air equivalent to 600 mg/cm² (about 5 meters) the true dose rate would be 4.3 milliroentgens-equivalent-physical per hour, which the film would measure as 3.2. For a 40-hour work week, the indicated dose would be 130 as compared with the true value of 170 mrep. Since the allowable exposure is 300 mrep/wk, this is not critical.

We have concluded from our experiments that the film method is reliable for measuring the beta radiation dose from uranium. It is not possible, from these studies, to predict results with other materials. However, the technique developed for determining the validity of the film method can be used for similar studies with other isotopes.

• • •

This article is based on a paper presented before the Radiation Division, American Industrial Hygiene Association, Cincinnati, Ohio, April 24, 1952.

The authors wish to acknowledge the assistance of Jerome Fleeman and F. S. Frantz, Jr., of the National Bureau of Standards X-Ray Section, in performing the monoenergetic measurements and of Sam Rothenberg, of this office, in processing the films.

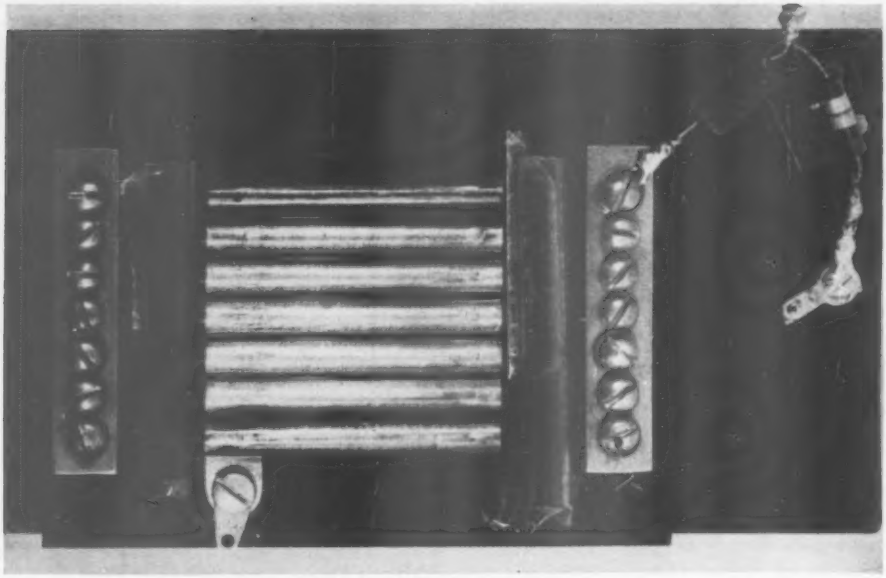

FIG. 1. Spark counter of the guitar type (see diagram in Fig. 6)

The Rosenblum Spark Counter

Two new designs for a wire spark counter make it possible to construct a cheap, rugged, low-background instrument for alpha counting. The counter operates at 3,000–5,000 volts, has marked directional properties, needs no amplifier, and is better than a scintillation counter in certain instances

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THE SENSITIVITY of electrostatic systems, consisting of a charged point or wire in close proximity to a plane, to the movement of ions has been well known for years. It is only recently, however, that work has been done to demonstrate the usefulness of discharge counters operated near their spark potential to detect heavily ionizing particles, such as alpha particles or protons. Previously, attention had been centered on radiation detectors operating in the Geiger or proportional regions of the discharge characteristics, whereas counters based on spark discharges were largely considered scientific curiosities and of little practical interest.

Early studies on discharge counters included the pioneer work of Greinacher (*1*) and his collaborators, especially Stuber (*2*), who investigated carefully the characteristics of the point spark counter, and of Chang and Rosenblum (*3*) who suggested a wire counter.

The main advantage of spark counters over other detectors lies in their speed of response and their simple geometry. These characteristics were exploited fully in the parallel plate counters developed by Neddermeyer (*4*) and by Pidd and Madansky (*5*, *6*). Such counters were employed as beta and gamma detectors by filling the space between the plane electrodes with a suitable gas, but this made their construction difficult and their operation uncertain. They have grad-

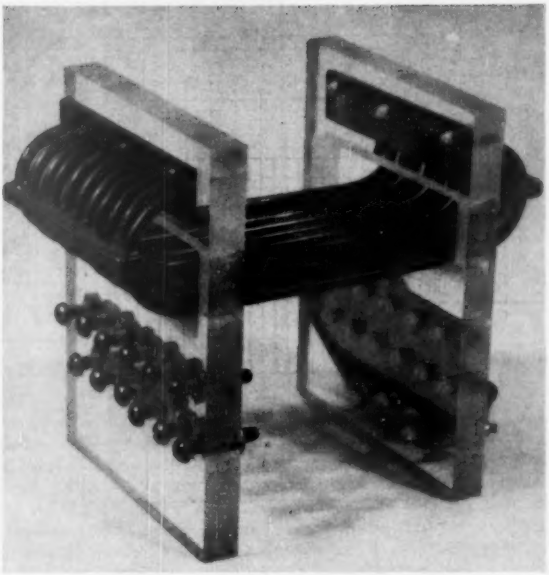

FIG. 2. Spark counter of the harp type (see diagram in Fig. 8)

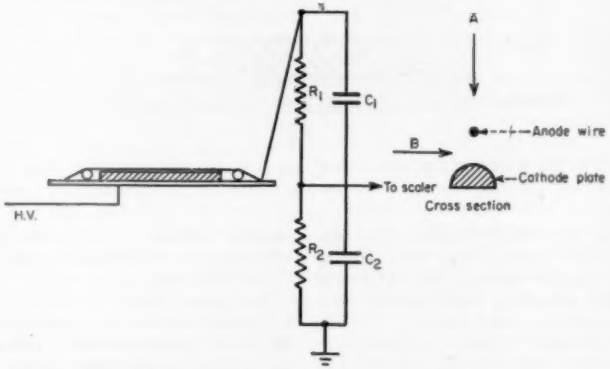

FIG. 3. Diagram of spark counter

ually been replaced by scintillation detectors, employing organic phosphors of short decay time.

The Greinacher point counter has too small a sensitive volume to be of serious practical interest. The Rosenblum wire counter, operating in air, does not suffer such limitation. Investigations have shown it to be a highly efficient and versatile fast detector of alpha particles (*7–10*).

This account is principally concerned with the design of Rosenblum counters for various applications. Figures 1 and 2 show two such designs, the "guitar" and "harp" counters, to be discussed later.

Basic Characteristics

In its most primitive form, the Rosenblum counter consists of a single, thin tungsten wire stretched parallel to a flat plate at a distance of 1–1.5 mm. A potential of the order of 3,000–5,000 volts is applied to the system so that a faint corona glow is visible around the wire. The passage of an alpha particle through the region between the electrodes will produce a spark discharge that can be quenched by means of a resistance-capacitance network.

Figure 3 illustrates schematically the main components of the apparatus.

The resistance network, R_1, R_2, is required to reduce the pulse amplitude to a value small enough to be handled by a scaler. The alignment of the wire can be carried out simply by watching the appearance of the corona.

Ricamo (*9*) and Payne (*11*) have investigated the effects of varying wire spacing and the extent of the alpha-sensitive region of the counter. Payne showed that the sensitive region is nearly coincident with the visible region of the corona; Ricamo demonstrated that the counter will operate with wire diameters ranging from 20–150 microns for flat cathode plates. When an alpha emitter is brought close to the counter, audible and visible sparks appear which are quenched by the *RC* circuit. For survey work, the scaler is unnecessary because the appearance or sound of the sparks give a good qualitative indication of activity.

In the experimental arrangement employed initially to investigate spark counters, curved cathode plates were used so as to localize the field and to permit the use of reduced working voltages. The cathode was a stainless steel rod, 6.5 mm in diameter, 6 mm long with rounded ends; the anode was a 0.003-in. tungsten wire stretched over glass rods. The distance between the electrodes was adjusted with mica spacers to 1 mm.

A potential of 3,000–4,000 volts applied to the system produced a visible corona along the wire with a steady corona current of 0.75 ma at 3,500 volts. R_1 and R_2 were 10^8 and 5×10^8 ohms respectively, and C_1 and C_2 25 and 2,000 $\mu\mu$f.

The spark counter is strongly directional and its characteristics were tested with particles incident along direction *A* (see Fig. 3), called by Connor (*7*) the "end-on array," and along direction *B*, the "broadside array." Figure 4 shows the voltage characteristics for both the broadside and end-on arrays. It is seen that for the broadside array a definite plateau exists once the unstable peak corresponding to a Geiger region is passed; no plateau was found for the end-on array, contrary to Connor's results.

The limited sensitive volume of the counter makes it suitable for range

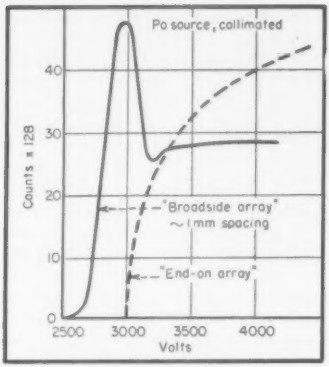

FIG. 4. Voltage characteristics of spark counter

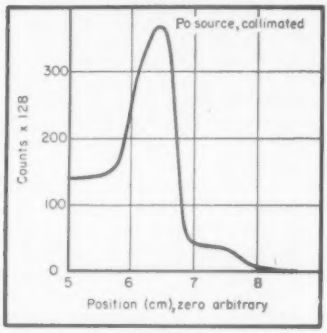

FIG. 5. Bragg range curve, spark counter

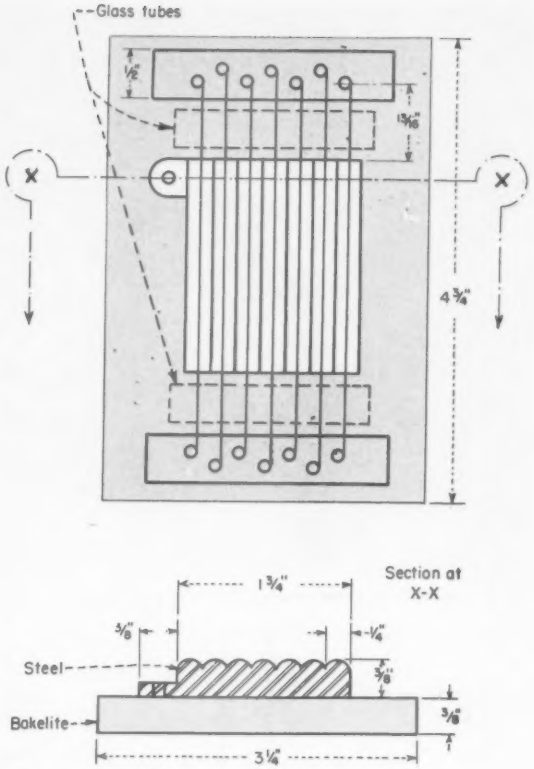

FIG. 6. Diagram of guitar counter

measurements on alpha particles from various sources. Figure 5 shows the Bragg curve obtained with a collimated polonium source in the broadside array. The straggling length is of the order of 2–3 mm, comparable to a good shallow ionization chamber, and could probably have been further reduced by shortening the length of the counter.

Multiple-Wire Counters

The main weakness of a single-wire counter lies in its very small sensitive volume which is confined to the corona region surrounding the anode wire. To increase the over-all sensitivity of the counter and its counting rate, it is necessary to increase the number of wires and place them as close together as possible.

Chang and Rosenblum found that this could be done by stringing a number of wires parallel to a flat plate, but they observed considerable interaction between the wires, leading to multiple sparks. They introduced thin separating walls between wires to overcome this difficulty.

Modified designs to operate such counters have been described recently by Connor (*12*) and by Guimarães and Sampaio (*13*). An alternative solution is that indicated in the "guitar" counter.

The guitar counter. Figure 6 is a diagram of such a counter. The cathode plate is ridged in such a way that each anode wire runs close and parallel to one of the crests of these ridges. The field is thus highly localized and interaction effects are avoided.

Figure 1 is a photograph of an experimental guitar counter. This counter is essentially a modified end-on counter possessing similar voltage characteristics. Because of the shape of its electric field and, hence, its sensitive volume, it is less sharply directional than the broadside counter. Figure 7 shows its Bragg curve.

The guitar counter has been used for many months as a qualitative detector of alpha activity for spotting, for instance, bench contamination or active areas in ore pieces.

By using it with the anode wires downwards, little difficulty has been encountered in keeping it clean. However, some contamination has been observed when assaying very finely ground powder samples, and this is ascribed to electrostatic attraction of the dust particles. This is also believed to be the correct interpretation of the phenomena observed by Kramer (*14*) in experiments with a point spark counter. As long as the counter is kept clean, background rates of less than 1 cpm can be attained.

The harp counter. To preserve the sharp directional response of the single

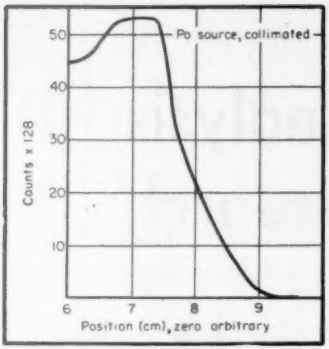

FIG. 7. Bragg range curve, guitar counter

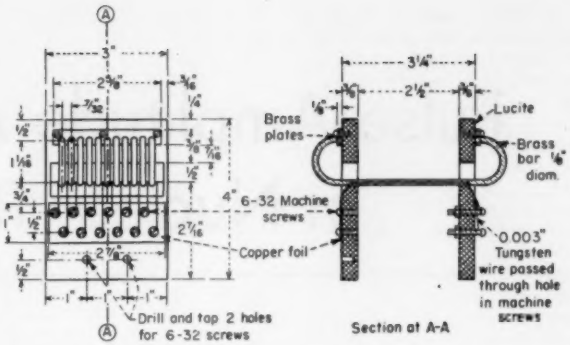
Brass plates
Lucite
Brass bar 1/4" diam.
6-32 Machine screws
Copper foil
0.003" Tungsten wire passed through hole in machine screws
Drill and tap 2 holes for 6-32 screws
Section at A-A

FIG. 8. Diagram of harp counter

wire counter, while multiplying the sensitive volume, a multiple-wire version of the broadside counter has been developed. This is called the "harp" counter and is illustrated in Fig. 8.

It consists of a number of wires strung between a set of smooth brass rods. The brass rods are curved at their ends to avoid sharp edges, which might cause spurious sparks, and the wires are carried over smoothly shaped Lucite supports for the same reason.

As the efficiency of the individual spark counter pairs is over 90%, the over-all efficiency is essentially proportional to the ratio of gap area to total counter area, as seen by a source above or below the counter. For the counter depicted, this efficiency is about 45%, and this could be further increased by using thinner brass rods.

Figure 2 shows the main features of a harp counter. Each wire can be tightened individually as in a string instrument; it is advisable not to tighten them too much to avoid mechanical oscillations when the counter is sparking.

Figure 9 gives two examples of range curves obtained with this counter and Fig. 10 its voltage characteristics.

Conclusions

Both harp and guitar counters offer several advantages over other types of alpha detectors. They are cheap, rugged, and easy to construct and to maintain. Experiments have shown that they are unaffected by illumination with strong daylight or ultraviolet light. The absence of any response to beta and gamma radiation tends to keep their background down to only a few counts per minute and makes elaborate shielding against cosmic rays unnecessary.

Since they operate in air, no elaborate preparation of the counters is required, and their large output pulse removes the need for amplifiers. To demonstrate the presence or location of alpha-active sources, observation of the sparks themselves is frequently sufficient, while, for more accurate work, the pulses can be fed directly into a scaler after suitable attenuation.

The spark counter has a faster response than a scintillation counter employing a zinc sulphide phosphor, but is about half as sensitive, area for area, because of its construction.

Its main applications lie, therefore, in qualitative measurements, in lecture demonstrations of alpha particles, and in range determinations of sources down to the order of one microcurie. Another possible application is for monitoring exhaust gases for alpha activity.

Altogether it seems reasonable to consider the Rosenblum spark counter a valuable tool in the nucleonics field.

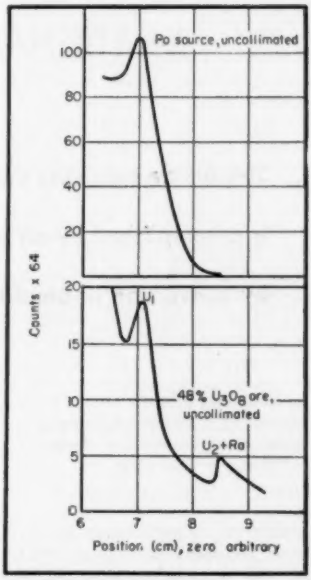

FIG. 9. Examples of range curves obtained with harp counter

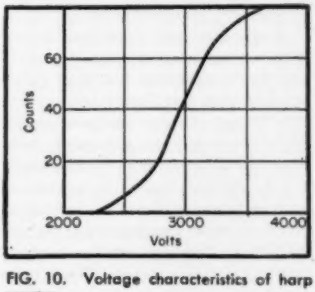

FIG. 10. Voltage characteristics of harp counter

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Pulse-Amplitude Analysis in Nuclear Research

PART IV

MULTICHANNEL ANALYZERS (Continued)

This article considers analyzers in which pulse-height analysis is accomplished by an expander-amplifier driving a chain of discriminators, by conversion of amplitude to time, and by information-storage devices

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FIVE TYPES OF multichannel analyzers were described last month in Part III of this four-part article. In this article, three additional categories of analyzers are described.

Expander-Discriminator

A succession of multichannel analyzers capable of a high degree of amplitude resolution has been developed by Bell and Kelley (*40, 41, 30*). The design of these analyzers results from the desire to determine, with considerable resolution in amplitude, the height distribution of a series of voltage pulses arriving at a very rapid rate. In this manner, data of high statistical accuracy can be accumulated in a few minutes. Short pulse durations, of about 1 μsec, short resolving times of a few microseconds, and many channels are thus necessary. An adequate amplitude resolution—for example, of 100 channels—should be accompanied by comparable stability of window width, about 1%.

In these analyzer designs, a biased, nonoverloading expander amplifier is so arranged that it views an adjustable portion—normally 10 to 20%—of the entire pulse-amplitude spectrum. This fraction of the spectrum, amplified, is then analyzed by a chain of 10 or 20 voltage discriminators.

The actual bias difference between successive discriminators is 3–5 volts, a value sufficient to insure small relative drift in window width. At the expander amplifier input, the windows appear smaller by the gain factor of the amplifier.

Successive portions of the pulse spectrum are analyzed by shifting the bias at the input of the expander. By changing the gain of the expander amplifier, the fraction of the spectrum observed by the discriminators can be altered.

As discussed in Part I, the bandwidth needed by an amplifier to reproduce faithfully the top of a short pulse is much greater than that needed to reproduce the entire pulse. The bandwidth of the expander amplifier must be wide indeed; in addition, it should be highly stable in gain and should overload gracefully; i.e., overload pulses should not cause baseline over- or undershoots.

To eliminate the need for unusually rapid pulse-height selectors which could respond to the short pulse of an expander amplifier, the burden of speed has been placed instead on a pulse-lengthener circuit located at the output of the expander amplifier. This circuit generates a flat-topped pulse of amplitude proportional to that of the expander-amplifier signal and of duration sufficient for the discriminators to register a count in the proper channel.

To the extent that these analyzers

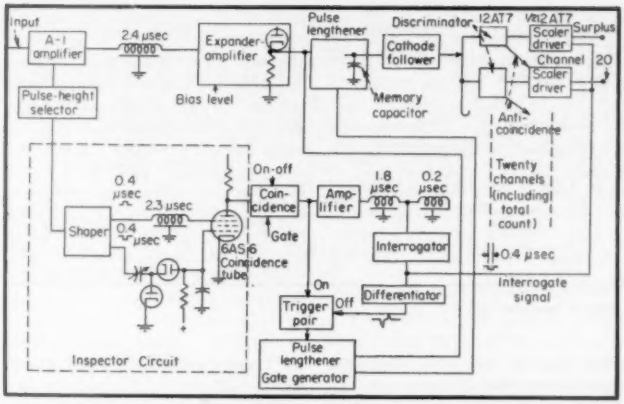

FIG. 31. Twenty-channel analyzer block diagram (*41*)

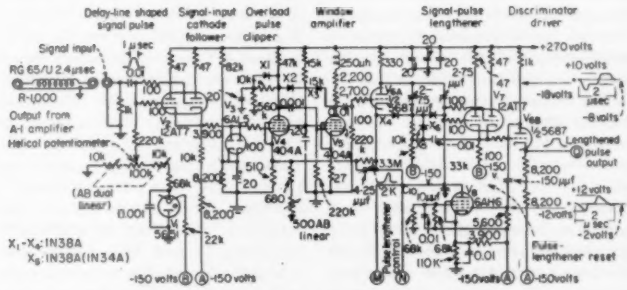

FIG. 32. Expander-amplifier and pulse lengthener for 20-channel analyzer (*41*)

contain a biased expander amplifier, a pulse lengthener, and a set of pulse-height selectors, their basic structure may be considered similar to that of the sorting-ladder analyzer (*36*).

A block diagram of the most recent model is shown in Fig. 31.* In addition to the expander amplifier, pulse lengthener, and discriminators, this 20-channel instrument† incorporates an "inspector" circuit to provide a fixed resolving time This circuit observes the short, negative, constant-amplitude pulses, or marker pulses, generated by the pulse-height selector in the Model A-1 amplifier. If two such pulses arrive within an interval of τ units of time—here chosen as 2.3 μsec—the inspector circuit rejects both pulses. Only those marker pulses preceded and followed by vacant 2.3-μsec intervals are accepted.

A marker pulse so accepted opens the pulse-lengthener gate, thereby permitting a signal pulse to be lengthened and sorted. (Note that a 2.4-μsec delay in the main signal path is required to insure opening of the pulse lengthener just prior to the arrival of the signal pulse.) The same marker pulse, delayed, generates an interrogating pulse which tests the 21 discriminators. Immediately thereafter, the pulse-lengthener gate closes, and the pulse lengthener is reset.

Adjacent discriminators are joined by anticoincidence diode connections. The interrogating pulse will thus generate an output signal in a scaler-driver tube when its discriminator has been triggered, but the discriminator in the next higher channel has not.

This analyzer design incorporates several significant improvements over earlier models. The earlier models, which contained no inspector circuits, gated the expander amplifier to prohibit a closely following input pulse from causing a false indication. The gating system made it possible for the expander amplifier to restore while a second (or third) signal existed at the amplifier input. If such a pulse had already passed its maximum, the analyzer would view an amplitude lower than the true value and register a count in the wrong channel. Gating in a signal channel is, therefore, considered unwise; gating is preferably carried out in accessory control circuits.

In these earlier models, an ingenious time-sequencing method of discriminator interrogation made anticoincidence connections unnecessary. The discriminators consisted of modified Schmitt circuits with biased type-6BN6 gated-beam-tube preamplifiers. The third grids of these tubes, starting with the highest channel, were each biased beyond cutoff by successively increasing amounts. A sawtooth interrogating signal applied to these grids thus tested each discriminator in turn. The first—and hence the highest—discriminator that had been triggered then generated an output pulse which not only tripped the appropriate scaler, but also terminated the interrogation and reset the pulse lengthener. The resolving time of the instrument thus depended upon pulse amplitude. However, operation of the trigger and sweep circuits associated with the discriminator chain was insufficiently rapid and reliable to prevent an occasional pulse from being recorded in more than one channel. This technique was, therefore, abandoned in favor of the present method.

A schematic diagram for the expander amplifier and pulse lengthener of this analyzer is shown in Fig. 32. The expander amplifier, of a design almost identical with that in the precision single-channel analyzer described in Part II, has a nominal gain of five. Thus, the 20 channels of 3 volts appear at the input, each to occupy 0.6 volt, or 72 volts for 120 channels. The 90-volt linear output spectrum of the A-1 amplifier is attenuated to approximately this 72-volt range by the 57 ft of RG65/U delay cable. As described previously, the expander includes overload features which allow the study of small pulses in the presence of large signals.

Crystal diodes X_4, X_5, and X_6, and tubes V_7, V_{8B}, and V_9 constitute the

* Figures 1–11 appeared in Part I of this article (NU, July '52, p. 20), Figs. 12–20 in Part II (NU, Aug. '52, p. 22), and Figs. 21–30 in Part III (NU, Sept. '52, p. 32).

† Manufactured by the Atomic Instrument Company, Cambridge, Mass.

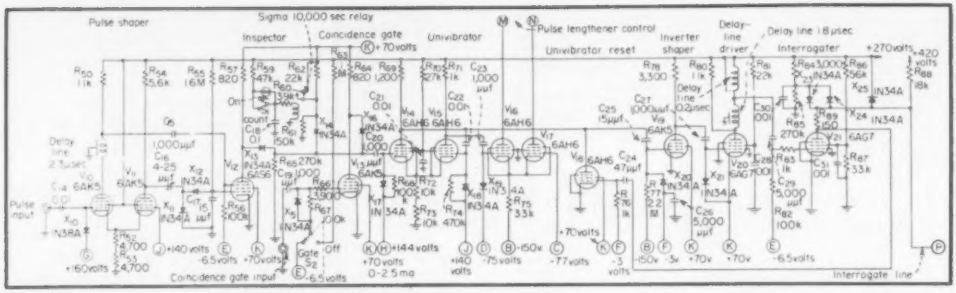

FIG. 33. Inspector and control circuits for 20-channel analyzer (41)

pulse lengthener (30). Quiescently, a pulse-lengthener control circuit (shown in Fig. 33) drains, via terminal M, 3.2 ma through X_4 and X_6. An inspected marker pulse opens the pulse lengthener by switching the constant-current drain from terminal M to N, the output of cathode-follower V_{6A}. Baseline disturbance due to opening of the pulse-lengthener is thereby prevented.

Capacitor C_{10}, the memory capacitor, is charged by the expander amplifier via cathode-follower V_{6A} and the series diodes to the amplitude of the signal pulse. When the signal pulse disappears, cathode-follower V_7 and diode X_5 hold the potential at the junction between X_4 and X_6 so that essentially all back potential appears across X_4. Less than 1.5 volts appear across X_6; since this diode is selected to have very high back resistance and to give the flattest top on the lengthened pulse, the reverse current is less than 1.0 μa, and the charge lost from C_{10} is negligible. The flat-topped pulse which results remains until 4.5 μsec after the arrival of the marker, when the pulse-lengthener control circuit again switches the constant-current drain to the cathode of X_6, thereby discharging the memory capacitor C_{10}.

Reset tube V_8, a tube which is normally cut off, hastens this discharge in the following manner. Because of the capacitive load presented by the discriminators to cathode follower V_{6B}, the signal appearing at its plate represents essentially the negative derivative of the lengthened pulse. During the discharge of C_{10}, this positive signal causes V_8 to conduct heavily and assist in the rapid discharge of C_{10}.

Were it not for this rapid discharge, a closely following low-amplitude signal might turn on the pulse lengthener again during the discharge of C_{10}, possibly causing thereby an output pulse of amplitude larger than that of the signal. Although the potential across C_{10} drops rapidly, the capacitive load of the discriminators causes a considerably slower decay in the lengthened-pulse output from V_{6B}.

The inspector and control circuits for the 20-channel analyzer are presented in Fig. 33. A 0.4-μsec marker pulse from the pulse-height selector of the A-1 amplifier is amplified and squared by tubes V_{10} and V_{11}. Such a positive pulse from the plate of V_{10} is delayed 2.3 μsec and applied to the third grid of V_{12}, a 6AS6 coincidence pentode. The negative marker pulse from the plate of V_{11} places a negative charge on C_{17}. The value of this capacitor is so adjusted that its potential, which rises exponentially toward +270 volts, crosses the zero axis 2.3 μsec later. If, however, a second marker pulse arrives during this interval, it recharges the capacitor, thereby delaying the zero crossing for at least an additional 2.3 μsec after its arrival, and causing rejection of the first marker, and of the second.

No output is obtained from coincidence tube V_{12} in the inspector circuit, unless the negative signal applied to the first grid terminates prior to the arrival, or during the presence, of the delayed marker pulse at the third grid. This requirement, in turn, is not met unless the marker pulse of interest is preceded and followed by vacant 2.3-μsec intervals.

A diagram of the time relationships between the signals in the inspector circuit is shown in Fig. 34.

The signal at the plate of V_{12} triggers a univibrator, V_{14} and V_{15}, which in turn opens the pulse-lengthener gate by transferring the constant current in V_{16} to V_{17}.

The leading edge of the univibrator pulse from the plate of V_{14}, amplified by V_{19} and V_{20}, is delayed 1.8 μsec. A 0.4-μsec line in the plate circuit of V_{20} "differentiates" the waveform; the positive 0.4-μsec signal which results is applied to the grid of interrogator-tube V_{21}. Crystal diode X_{25} maintains the quiescent plate potential of this normally cut-off tube at +271 volts; the arrival of the positive grid signal develops on the interrogator line a negative square pulse, the lower level of which is held at +265 volts by diodes X_{23} and X_{24}. Careful control of the potential limits of this pulse with respect to +270 volts is necessary for proper operation of the scaler-driver circuits actuated by it.

A similar signal from the plate of V_{21}, differentiated by C_{24} and R_{74}, is applied to univibrator-reset tube V_{18}. The positive trailing edge of this pulse drives V_{18}, which resets the univibrator, and closes the pulse-lengthener gate, thereby restoring all control circuits to quiescence. Because of the 1.8-μsec delay and the 0.4-μsec pulse width, the duration of the pulse-lengthening gate is 2.2 μsec. The time relationships between these control waveforms are shown in Fig. 34.

The discriminator-, anticoincidence-, and scaler-driver-circuit combinations, portrayed in Fig. 35, are conventional in nature, but have been engineered to require a minimum number of tubes. Instead of a Schmitt circuit, a capacitively-coupled regenerative clipper (V_1, V_2, V_3, and the discriminators of the other channels) is used for pulse-height selection. A positive output pulse from the right plate of one of these tubes raises the grid of the following scaler-driver tube (V_{2A}, V_{4A},

V_{6A}, and the scaler drivers in the other channels) thereby permitting it to conduct and generate a negative output signal at its plate when an interrogating pulse appears at the cathode. If the discriminator in the next higher channel is triggered, however, its left plate delivers a negative anticoincidence signal via a crystal diode (X_4, X_8, and the similar diodes in the other channels) to suppress the scaler-driver grid signal, so that this tube remains cut off when interrogated. Note that the interrogation technique used obviates the need for storage of anticoincidence information as discussed in Part II.

Several circuit features in the discriminator are of interest. The grid of the normally conducting triode is returned via a crystal diode (X_3, X_{11}, and the similar diodes in the other channels) to a reference potential of +100 volts. After a triggered interval, this diode rapidly restores any charge lost by coupling-capacitor C_4, C_7, C_{10}, and the coupling capacitors in the other channels; disturbance of threshold is thereby minimized. Breakaway-diodes X_2, X_6, X_{10}, and those in the other channels, and compensating-inductances L_1, L_2, L_3, and those in the other channels, insure an abrupt leading edge for the negative anticoincidence signal.

The input grids of the discriminators are connected via high-back-resistance crystal diodes, X_1, X_5, X_9, and those in other channels, to points of successively lower bias potential, each separated nominally by 3 volts. Exact adjustment of window width is rapidly accomplished by means of vernier rheostats between adjacent potential taps on a voltage divider. In this manner, a decrease in width of one window is absorbed by a slight increase in width of all other windows, and not entirely by the adjacent window.

The 20-channel analyzer employs 7 tubes in the expander amplifier and pulse lengthener, 12 tubes in the inspector and control circuits, and only 1½ tubes for each channel (including the discriminator, anticoincidence, and scaler-driver circuits, but excluding the scaler). The reliability of operation should, therefore, be relatively high.

A fixed resolving time of 2.4 μsec results from the nature of the inspector circuit. The analyzer will properly sort pulses having a rise time as short as 0.2 μsec. Since all discriminator potentials, including heater-supply voltages, are regulated, the stability of window width is expected to be of the order of 1%. With a single measurement, 20-channel data are obtained; through only one adjustment of expander bias, these 20 channels can be located to observe any portion of a pulse-amplitude spectrum. Hence, only six repeated measurements are necessary for 120-channel resolution.

Amplitude-to-Time Conversion

Two pulse-height analyzers, developed during the past few years, measure pulse amplitude by converting input pulses of various amplitudes into pulses of duration proportional to input amplitude. Since time is a dimension which can be measured most accurately, the ultimate accuracy of a time-conversion analyzer depends most upon the quality of the conversion from amplitude to time. By determining the boundaries of the various channels with a mechanism common to all channels, one can substantially eliminate the problem of relative boundary drift.

A 99-channel analyzer, suitable only for slow counting, has been described by Wilkinson (*42*). In this analyzer, the input pulse charges a storage capacitor to the pulse height; this capacitor is then discharged at a constant rate by a pentode connected as in a Miller time-base circuit. The rectangular gate generated during this discharge, which has a duration proportional to pulse amplitude, gates a free-running oscillator and so produces a pulse sequence of number proportional to input amplitude.

This number is counted by two cascaded rings-of-10, the first determining the units digit and the other, triggered once for each cycle of the units ring, the tens digit of the number of pulses. A series of 20 relays, each actuated by a triode pair in the two rings-of-10, energizes a 10-by-10 matrix of telephone-message registers. Thus, a count of 57 is recorded by connecting one terminal of the 57th register to ground through the relay actuated by the seventh tube in the units ring.

The other register terminal is connected, via a relay associated with the fifth tube in the tens ring, to a tube which passes a heavy current pulse after the rings have accepted the count. A set of diodes, of which one

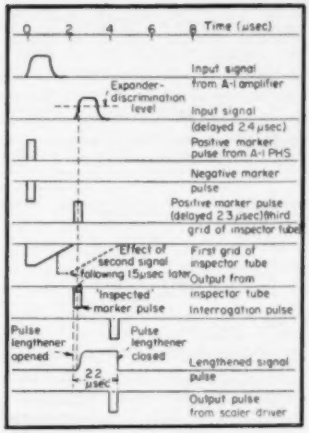

FIG. 34. Time diagram for 20-channel analyzer (*41*)

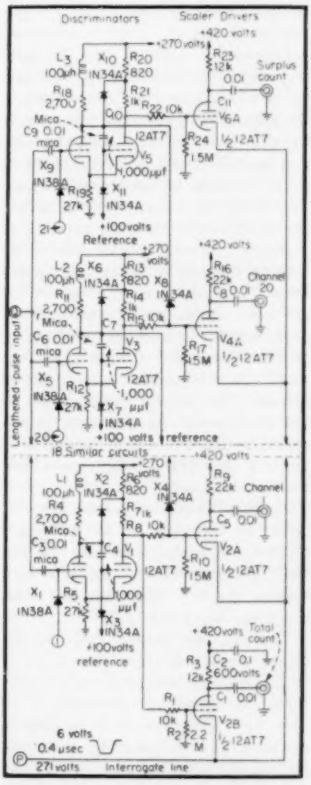

FIG. 35. Discriminator and scaler-driver circuits for 20-channel analyzer (*41*)

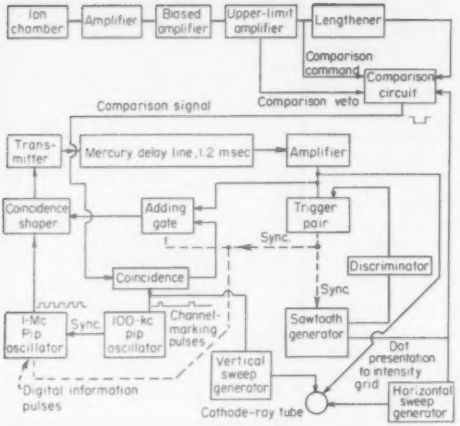

FIG. 36. Block diagram of analyzer that converts amplitude to time (43)

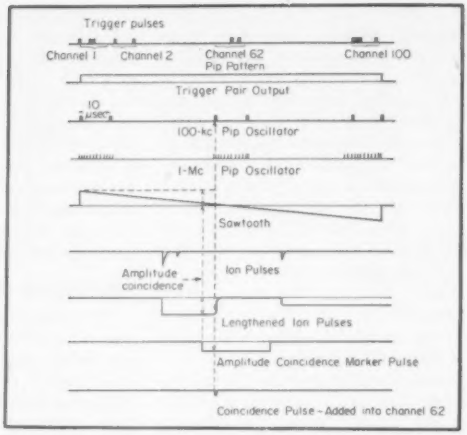

FIG. 37. Time diagram for amplitude-to-time-conversion analyzer (43)

is connected in series with each register, prevents the current pulse from finding devious parallel paths through the register matrix.

Because of a dead time in each register of 0.1 sec, any register can never count faster than 10 per second. To prevent an intolerably high counting rate from jamming a register and causing spectrum distortion, each input pulse is made to initiate a blocking time of 0.1 sec during which no subsequent pulses can enter the system.

Auxiliary circuits are necessary to perform a series of supporting operations. An initial 40-msec period is provided during which the 27-Mc oscillator is gated, and the number of cycles so produced is counted by two rings. A subsequent 20-msec period permits the large current burst to record a count on the appropriate register. During a third interval of 3 msec a high-speed relay resets the two counting rings.

Pulses of amplitude greater than 99 units are counted by a "surplus" register. A high-speed relay diverts the registering current pulse to this surplus register whenever the tens ring completes a full cycle.

The analyzer uses a total of 70 tubes, of which 40 are contained in the two rings-of-10 and relay-driver circuits. In speed of operation, it is very slow; its 0.1-sec resolving time is exceeded only by that of the mechanical analyzer described previously. On the other hand, with only one measurement, it provides 99-channel information. The calibration curve shows no deviation from linearity greater than 1%. Long-term drift, likewise, is equally small.

A second pulse-height analyzer using amplitude-to-time conversion is one designed by Hutchinson and Scarrott (*43*). This unit, developed in an effort to increase reliability by reducing the complexity of multichannel analyzers without any sacrifice in number of channels, storage capacity per channel, or accuracy, accumulates pulse-height data in a memory device, a supersonic mercury delay line.

Input pulses, after being amplified and lengthened, are compared with a linearly rising sawtooth. The time required for the sawtooth to reach the amplitude of the lengthened pulse is proportional to the pulse amplitude. After this conversion process, information concerning pulse amplitude—in time-duration form—is stored in the mercury delay line, through which information recirculates continuously with a period of 1,200 μsec. This interval is divided into the appropriate number of consecutive channels.

Within each channel, the number of pulses stored is indicated in binary code by a sequence of 1-μsec pulses. For example, in a 120-channel system, 10 μsec are available per channel, so that 10 short pulses can be stored 1 μsec apart. The maximum number that can be so represented in binary form is 1,023. A block diagram for this system is shown in Fig. 36.

Stored pulses circulate through the mercury delay line, the output amplifier, the adding gate, and the coincidence-shaper stage in the transmitter. Information in the line exists as amplitude modulation of a 13-kc carrier. The trigger pair, which is actuated once each cycle by an initiating pulse circulating in the line, starts the linearly rising sawtooth. In the comparison circuit, a comparison pulse is generated at the instant of amplitude equality between the sawtooth and a lengthened input pulse. The comparison pulse permits a coincidence circuit to pass the following channel-marking pip from the 100-kc oscillator. The channel-marking pip, combined with the 1-Mc pips from the digital-marker oscillator in the coincidence-shaper stage, acts to increase by one the binary number stored in that channel interval. A timing diagram which clarifies the relationship between these signals is portrayed in Fig. 37.

Data are presented on a cathode-ray tube in which the spot is swept horizontally once each 1.2 msec and swept vertically once per channel interval, thereby forming a raster similar to a television pattern rotated 90 degrees counterclockwise. The sweep, dimly visible, is intensified by each pulse arriving from the delay line. Columns of dots are produced, as shown in Fig. 38; each column represents a different channel. The vertical array of dots in each column is a binary representation of the number of pulses within that channel. Channel 1 shows, in binary notation, 11101 pulses accumu-

lated. Transferring this to decimal notation, the number is $2^4 + 2^3 + 2^2 + 2^0 = 16 + 8 + 4 + 1 = 29$ pulses.

Thus, the contour formed by the tops of the columns forms an approximate logarithmic measure (to the base 2) of the distribution function. One is able to observe the distribution grow as data are collected. When sufficient data are accumulated to reduce statistical variations to an acceptable level, the measurement is terminated, and the distribution is recorded.

Using only 82 tubes, excluding the power supply, this analyzer is a versatile unit which, in its present form, is capable of 60-channel resolution with a storage capacity of about 10^6 counts per channel, 80-channel resolution with a capacity of 32,000 counts per channel, or 120-channel resolution with a capacity of 1,023 counts per channel.

Since the average time required to record a pulse is half the period of the mercury delay line, the maximum counting rate of the present machine is about 1,600 per second. Here, again, a common mechanism has been used to determine the boundaries of all channels. Hence, the uncertainty of channel width and the deviation of the response from linearity can both be as small as one part per thousand.

Other Storage Techniques

Many other techniques for determining pulse-height distributions based upon information storage in a memory device have been proposed and used.

One proposal for an analyzer which would accumulate and display data continuously involves a secondary-emission, beam-deflection, amplitude-analyzer tube (*44*). This storage-tube, designed for studies of continuous functions—such as those in speech analysis—has not been developed to a point where it is capable of the necessary amplitude resolution.

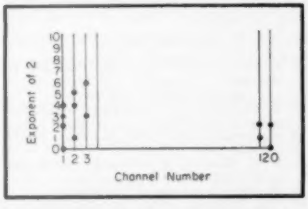

FIG. 38. Data presentation on cathode-ray tube in amplitude-to-time-conversion analyzer (*43*)

Another proposal consists of converting pulse-amplitude information into time-duration form in a manner similar to that of Wilkinson (*42*). The sequences of oscillator pulses by which amplitudes are defined are stored on magnetic recording tape for analysis with a scaler-register matrix.

Photographic Methods

Part I of this series described the single-frame, time-exposure technique of Maeder (*3*), in which a single frame of film is exposed to lengthened signal pulses displayed repeatedly on the face of a cathode-ray synchroscope. On the resulting film record, the density of exposure at a given height—usually measured with a densitometer—thus corresponds to the number of pulses having that amplitude. This technique has been used with great success by Campbell and others (*45–49*).

The amplitude distribution can be observed in diffuse outline directly by viewing the exposed film through an optical wedge or by rephotographing the original exposure on high-contrast film through an optical wedge. Because of the nonlinear characteristic relating film density to light exposure, the ordinate values of a distribution curve so obtained are not accurate. For accurate determinations of these ordinate values, one can compare with a densitometer the density of exposure of the single frame with the density of a calibrated reference frame on which the density of recording—in pulses per unit amplitude—is made a linear function of amplitude.

For applications in which the resolution available from a single frame is inadequate, use has frequently been made of a moving-film technique, in which a separate record of each pulse is made on a continuously moving film.

Stone and Borkowski (*50*) have described one method, in which a film, moving continuously in the x-direction past the screen of a cathode-ray synchroscope, views a series of vertical lines with heights proportional to pulse energy. These lines, as indicated in Fig. 39, are produced by pulse signals applied to the y-deflection plates, while the x-position of the beam is left undisturbed. The "integral-bias" data contained on the film are reduced by scanning one channel at a time either visually or with an automatic photoelectric scanner.

Deutsch and others (*51–54*) have used a similar technique of moving-film analysis, but with the data recorded in the form of spots. The distance from each spot to a baseline parallel to the film edge is proportional to pulse amplitude. In this method, the dots can be produced by intensifying the cathode-ray beam at the peak of a lengthened pulse or by viewing the synchroscope through a mask containing a slit which crosses the pulse outline at its peak. More data can be stored per foot of film using dots rather than a line presentation, because a dot occupies only one channel at a time. Photoelectric scanning of the dots on the film can be employed to reduce the data into useful form.

A single-channel pulse-height analyzer described recently by Wright (*55*) makes use of a somewhat similar cathode-ray presentation in which the instantaneous pulse potential is applied to the x-deflection plates, and the time derivative of pulse potential is applied to the y-plates. On the Lissajous pattern so created, the distance from the origin to the crossing of the horizontal axis is then a measure of pulse amplitude. Instead of recording this distance on film for each pulse, the analyzer utilizes a multiplier phototube to observe with a slit a selected portion of the horizontal axis and determines with a ratemeter the number of pulses per unit time having amplitudes within this slit. By plotting slowly, on a second oscilloscope, the counting rate as a function of horizontal position of the slit, one can get a curve of pulse-height distribution.

Photographic techniques all involve simplified equipment, both to store information on the film and to scan the film photoelectrically for data reduction. However, this resulting simplicity, and hence, reliability, is obtained at the expense of speed—at a cost of longer time required, not only for processing of data, but also for

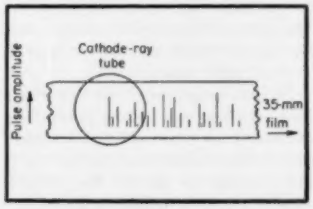

FIG. 39. Data storage pattern on 35-mm film in moving-film analysis (*50*)

processing of the film. On the other hand, data are stored permanently, always available for re-analysis to study carefully at a later time any feature of the spectrum. Additional versatility exists in that the film may be scanned with any desired number of channels (consistent with adequate statistical accuracy within each channel) and any window width or distribution of window widths.

Photographic methods of analysis, in which data from a cathode-ray tube are recorded on moving film, are subject to some of the same considerations as are beam-deflection analyzers, namely, the concept of a preamplifier driving one or more discriminators. Gain-bandwidth product and stability limitations similar to those discussed in Part III enter not only in the process of recording data on the film, but also in reducing it from the film.

The problem of determining whether or not a spot barely lies within a given channel on a film is similar to the problem of deciding whether or not a cathode-ray beam which grazes a target electrode actually strikes it. The data-reducing process must, therefore, include a discriminatory operation.

Another problem is that of choosing an economical rate of film motion relative to average pulse rate, yet one in which loss of information due to overlapping spots is not excessive. It appears, with presently available spot diameters and film sensitivities, that sufficient data can be recorded on 100 ft of 35-mm film to obtain 50-channel resolution with statistical uncertainties of about 2%.

Conclusion

In the design of a fast pulse analyzer having high amplitude-resolving power and stability, several points are of importance. To obtain high amplitude resolution, it is desirable to sort pulses at a high level; i.e., the discriminator circuits should preferably be preceded by some amplification. The gain of these amplifier stages must be highly stabilized if the apparent window width is not to vary.

To obtain high speed and to prevent false response to overload pulses, the amplifiers and associated discriminators should incorporate nonoverloading features to permit the circuits always to return rapidly to quiescence after the occurrence of a pulse. In addition, arrangements to prevent baseline overshoot at the end of an overload pulse are desirable. Scaling circuits must be capable of a time resolution corresponding to the highest channel counting rate.

One of the major problems in a multichannel analyzer is the large number of elaborate and complex circuits which require proper adjustment and maintenance. The larger the size of an analyzer, the greater the probability of equipment failure and the lower the reliability of operation and of the data presented.

Indeed, many of the more complex analyzers incorporate monitor or calibrating units which permit rapid test of the system's behavior. Others provide pertinent test points where a check on voltage and waveform can be used to diagnose the cause of any improper operation.

Data from a multichanne pulse analyzer can be relied upon only when obtained from a system in which monitoring tests are carried out frequently, or in which breakdown or maladjustment is almost negligible.

Obviously, any simplification of the circuits pertaining to an individual channel of a multichannel pulse analyzer results in a considerable improvement and economy, because the simplification applies to all channels.

The use of analyzers in which a common mechanism determines the boundaries between various channels is, of course, an aid to simplicity, reliability, and stability. Amplitude-to-time conversion and photographic-type analyzers enjoy this advantage.

In conclusion, it may be well to list some of the desirable characteristics of a multichannel analyzer, as outlined by Wilkinson (*42*).

Number of channels. The greater the degree of amplitude resolution, the greater need be the number of channels or potential number of channels.

Stability. The accuracy with which a distribution function can be determined is no better than the constancy of channel width. Relative drift in channel boundaries is thus highly detrimental. A small drift common to all boundaries, if but a fraction of channel width, is not of serious consequence.

Speed of operation. For rapid determination of amplitude distributions with low statistical uncertainties, the analyzer must be able to accept random pulses at high average rates of arrival. The resolving time is, in part, a measure of speed.

Simplicity. For reliable data, the construction and operation of an analyzer must be as simple as possible. One measure of simplicity is the total number of tubes divided by the number of channels.

Linearity. For convenience, an analyzer should have equal channel widths, with calibration as nearly linear as possible. Stability is more greatly to be desired, however, than equal channel widths or linearity.

Universality. A pulse-amplitude analyzer should respond to pulses over as wide as possible a range of amplitude, duration, rise time, and shape.

Having in mind these requirements, as well as the specialized needs of a particular application, the reader may himself evaluate the merits of the various analysis systems described.

* * *

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OTHERS IN THIS SERIES . . .
were articles on
Voltage discriminators
Single-channel differential analyzers
Multichannel analyzers—I
which appeared in July, August, and September, respectively.
This is the last in the series.

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A Comparison of

Iodine-131 Counting Methods

Data on the comparative efficiency and sensitivity of nine methods of counting the betas and gammas of I^{131} are presented as a basis for choosing an assay method in keeping with a given set of conditions and requirements

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SELECTING the method and equipment for iodine-131 counting, with the requirements of a particular problem in mind, can lead to a choice of one of several more or less standard procedures. As is true of radioassays in general, each of the methods discussed in this over-all comparison is, in varying degree, a compromise between technical limitations, economics, and a theoretically ideal method.

Ideally, the sample should be simply and quickly prepared without loss from adsorption on the walls of the containers or from volatilization. The detecting apparatus should be stable and hold its calibration for at least the six months between shipments of NBS standards; it should have a wide range of sensitivity so that it can match the range of activities encountered, without need of concentrating or greatly diluting the sample.

The chemistry of iodine complicates handling I^{131} because it can be lost from acidic solutions. Thus, the conventional dry and wet ashing procedures may give unpredictable losses. Concentrating I^{131} solutions by means of heat or air drying was found by Kelly *et al.* (*1*) to result in appreciable losses that were progressively greater from plasma, urine, and distilled water. This tendency to loss following oxidation to iodine should be, but is not always, combatted by use of alkali and an anti-oxidant such as $NaHSO_3$ in concentrations of 0.005–0.5*M*.

Routine addition of carrier NaI to the assay sample is highly desirable, although many biological samples normally will contain sufficient quantities of iodide to act as carrier. The I^{131} shipped from Oak Ridge contains $NaHSO_3$, but ordinarily no carrier iodide.

While the exact sensitivity and efficiency of the different kinds of assay systems will vary from one laboratory to another, the ratings given here should, in general, hold. However, variations in details of geometric arrangement and sensitivity of components can cause minor shifts in the ratings.

As discussed later, Marinelli and Hill (*2*), in describing a 2π Marinelli counter, noted that it counted with an efficiency of 0.033% for I^{131}, while the 4π "Texas" G-M counter had an efficiency of 0.80%. Oshry and Schmidt (*3*) reported an efficiency of 0.02% for a Marinelli counter; this was but slightly better than the 0.017% efficiency obtained when a cup of liquid was placed beneath the sensitive volume of the counter tube mounted horizontally. Freedburg *et al.* (*4*) noted that the Marinelli system had a limit of sensitivity of 0.25 μc (about the same as the preceding), whereas a dry mount of I^{131} counted by a mica end-window G-M tube had a limit of 0.001 μc.

Comar (*5*) observed that a mica end-window tube counted a dry film of I^{131} about four times more effectively than did a 30 mg/cm^2 glass-wall cylindrical tube mounted horizontally at the same distance above the film. Both gave higher counts than a Bale-Haven dipping tube used to count the same amount of iodine in solution.

A dry film of I^{131} proved the most efficient sample mount for an end-window beta counter. The film was about 30 times more effective than when the I^{131} was dissolved in water and presented as a flat disk 25 mm in diameter and 4 mm deep (infinite thickness for I^{131} betas in water is 2.2 mm). The latter, in turn, was some 8.5 times more effective than when it was in a disk 90 mm in diameter by 7 mm deep.

By increasing the geometry to about 50% and reducing absorption, the recently developed windowless gas-flow counters offer greater sensitivity than the standard mica end-window counters. The increase is as much as 5–500 times, depending on radiation characteristics of the isotope being counted. In the case of I^{131}, Graf *et al.* (*6*) found a sensitivity relation of 0.13 to 1.0 to 1.2 for a mica end-window G-M tube, a windowless flow G-M counter, and a windowless flow proportional counter, respectively. Samples

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Relative Effectiveness of Nine Methods of Detecting Iodine-131

Method and sample preparation	Container	Volume of sample* (ml)	Background (cpm)	Net cpm	Efficiency (%)	Minimal detectable I^{131} (μc)	Maximal volume of sample (ml)
1 Marinelli 2π counter		5.0	36	287	0.166	0.01	40
(Liquid samples)							
2 "Texas" 4π counter		0.4	306	1,106	8.01	0.002	75
(Liquid samples)							
3 High-pressure ion chamber (4π)							
(Liquid samples)							
"Rate of drift"		25.0	93†	110†	Low	~0.1	200
"Electrometer"		25.0	10†	40–44†	Low	~0.1	200
4 Quartz-fiber electroscope	Porcelain	1.0	0.068†	2.65†	High	0.0008	10
(Dry samples)	Steel	1.0	0.061†	2.10†	High	0.0009	3
5 Mica end-window G-M counter	Porcelain	1.0	38	648	1.88	0.001	10
(Liquid samples)	Steel	1.0	27	353	1.02	0.0013	2.5
6 Mica end-window G-M counter	Porcelain	1.0	38	1,775	5.14	0.00033	10
(Dry samples)	Steel	1.0	28	1,699	4.93	0.00026	2.5
	Top shelf		28	3,909	11.3	0.00011	
7 Scintillation counter	Porcelain	1.0	260	98	0.28	0.042	10
(Liquid samples)	Steel	1.0	229	60	0.17	0.061	2.5
8 Scintillation counter	Porcelain	1.0	260	65	0.19	0.62	10
(Dry samples)	Steel	1.0	229	60	0.17	0.61	2.5
	Very close		229	268	0.78	0.13	
9 Gas-flow proportional counter	Porcelain	1.0	105	9,762	28.3	0.00016	10
(Dry samples)	Steel	1.0	45	8,953	25.9	0.00008	2.5

* An NBS standard containing 34,510 disintegrations per minute per milliliter.
† This figure is divisions per minute (not cpm) as given on instrument scale.

for the last two systems must be completely dry, since water vapor seems to poison the counting gases.

Methods and procedure. A National Bureau of Standards sample of I^{131} was diluted volumetrically so that the final solution contained 34,510 dpm/ml at a specified time. The walls of the flask had been wetted with 2.0 ml of 0.05*M* NaI and 2.0 ml of 0.5*M* $NaHSO_3$, followed by 200 ml of water, before adding the NBS standard. The solution was adjusted to *p*H 10 by NaOH. The samples were prepared from this solution using micropipettes or doubly-tested transfer pipettes.

Where necessary, this standard was diluted with distilled water. The samples that were subsequently dried by infrared lamps for comparison of liquid versus dry counting were without weighable residue and were, therefore, considered to exhibit no self-absorption.

The containers for these comparison samples were (*a*) stainless steel cup planchettes, 24.5 mm internal diameter by 7.2 mm deep with a surface of 4.71 cm^2, and (*b*) Coors porcelain ashing capsules 40 mm internal diameter by 11 mm tall with 12.57 cm^2 surface. The capsules had been coated with "liquid bright platinum" and fired to provide a rugged electrical conducting surface. Without the coating, progressively lower counting rates are obtained in gas-flow counters due possibly to accumulation of residual charge. These capsules have two disadvantages in that their floors are not completely plane and they have an intrinsic background. On the other hand, the entire ashing and counting procedure can be carried out in this one vessel.

All values reported in the table above are the average from triplicate samples prepared and counted in exactly the same way. The same liquid and dry preparations were used in methods 4 through 9.

The samples for the self-absorption curves were prepared by slowly evaporating constant volumes of mixtures of the standard and varying amounts of urea.

With two exceptions, the components of the different counting systems were standard equipment (*7*). They were used according to procedures now generally accepted (*8, 9*). The samples were counted so that the standard deviation of the total number of counts was ±1.5% or less, except in one series where it was ±1.9% or less (*10, 11, 12*). The time distribution between samples and background was either optimal or in favor of excess background count. The counting rates were low enough not to require correction for coincidence.

The essential results are collected in the table above. The column "efficiency (%)" is the standard expression: [(cpm/ml ÷ dpm/ml) × 100]. The "maximal volume of sample" relates particularly to the first three wet-counting methods; it indicates the largest volume of a liquid that can be counted in that particular counting system. This be-

comes an advantage in that the use of large volumes of low-specific-activity liquids, such as urine, can give the same effect as an increased sensitivity of the apparatus.

The minimal activity detectible is defined here as the *net* counting rate equal to the background. This number divided by the observed efficiency and 2.2×10^6 gave the values in the "minimal detectible I^{131} (μc)" column. A sample counting rate of twice the background counting rate is the smallest ratio consistent with good statistics, assuming total sample counts of at least 5,000 and usually 10,000.

Each of the procedures and methods in the table is described in what follows:

Marinelli 2π Counter

The arrangement of apparatus was essentially that described by Marinelli and Hill (*2*) except that (*a*) a bismuth wall G-M tube was used which was some five times more sensitive to I^{131} gamma energies than the ordinary silvered-wall tube (*13*); (*b*) the G-M tube was not protected by a brass sleeve; and (*c*) the 40-ml sample beaker, which accommodates the tube through its base, was designed (and locally manufactured of 35-mm OD and 48-mm OD Pyrex tubing) to give a cuff of liquid 65 mm long, which is approximately half the length of the cathode of the G-M tube. The 40-ml volume was a compromise between the higher counting rates obtained by using additional amounts of a constant specific activity liquid in a larger cup, e.g., 250 ml of the same urine sample, and the lower counting rates which would result if a small, fixed amount of active material, such as a tissue sample, were diluted to 250 ml.

The reproducibility of the method is very good. The efficiency of counting is low, but the system is highly stable and holds its calibration for at least six months of steady use. Its special usefulness is in counting urine, where a wide range of activity is encountered. If the activity in the urine is low, the cup is merely filled to the 40-ml line from a known volume of urine. When the urine is very active, a measured sample is taken and water added to the 40-ml mark.

"Texas" Counter

This 4π Geiger counter (Fig. 1) is similar to that referred to by Marinelli and Hill (*2*).

High sensitivity to gamma rays is secured by means of a long anode, large sensitive volume and walls of high-atomic-number materials. The anode is arranged as a series of wires connected in parallel surrounding a well 12.5 cm deep and 3.8 cm in diameter into which the sample is placed. Its background was 280 to 320 counts per minute with four inches of lead on all sides and about 1,800 without shielding. The most reliable counting range, therefore, would be between about 600 and 4,000 cpm.

The location of the active source in the well influences the counting rate so that all samples should have constant geometry. This means that the sample should be a liquid.

FIG. 1. "Texas" counter with plastic sleeve-protector partly lifted out of center well. Lusteroid sample-holder tube has been inserted into sleeve

The counting efficiency proved much higher than that for the other gamma-detecting methods and compared favorably with the average beta-ray counter. Although it is often necessary to make several dilutions before obtaining one in the optimal counting range, the same is frequently true in preparing wet and dry samples for the beta counters. The minimal detectible microcuries, 0.002, can probably be further reduced by better shielding and by locating the counting setup where there is less stray radiation.

A 10- or 20-ml liquid sample in a 100-ml "Lusteroid" tube has proved very satisfactory. Increased counting rates can be obtained by using additional amounts of a constant-specific-activity liquid. The counter is sensitive to variations in geometry of the sample, so that a standard procedure should be established.

High-Pressure Ion Chamber

This ionization chamber was a locally manufactured variation of a more-or-less-standard 4π instrument (*14, 15*) used at many AEC laboratories. See Figs. 2, 3, and 4.

The tank was filled to only 100 lb/in.^{2} with argon to minimize its background sensitivity and oscillation from vibration of the collectrode. A vibrating-reed electrometer plus Brown recorder was used to detect the current which was recorded either as a rate-of-drift or as an electrometer reading with a range of six orders of magnitude. The system has excellent day-to-day stability according to regular checks using permanent gamma-ray standards.

Since the response was linear with respect to the activity of the I^{131}, the scales were calibrated by use of known activities contained in specified volumes; accordingly, the efficiency has not been calculated. The estimate of sensitivity in the table was based on the limit of distinct differences on the trace of the Brown recorder. If additional sensitivity were sought by use of larger volumes of a constant-specific-activity sample such as urine, calibrations at that particular volume would have to be established. The chamber response is independent of geometry of the sample only at the very bottom of the well, and then only to a limited degree. In practice it is much simpler to keep sample geometry and volume constant.

Unfortunately, high-pressure ionization chambers are not commercially available, despite their many desirable features.

Quartz-Fiber Electroscope

A conventional Landsverk electroscope with removable end was used. The dry samples were simply placed inside the electroscope chamber below the collecting electrode so that a geometry of slightly better than 50% was obtained. The measurements should be carried out in a constant-humidity thermostatted room and the day-to-day sensitivity checked and corrected for, if necessary, by use of an appropriate permanent standard. The electroscope has proved remarkably stable considering that the in-

FIG. 2. On right is wheel-mounted, heavily shielded high-pressure ion chamber, constructed from steel oxygen bottle. Shield has successive layers of 1-in. iron plate, 1-in. lead plate, and 6 in. of high-density concrete. Output is amplified by vibrating-reed electrometer (center) and read on Brown recorder (left)

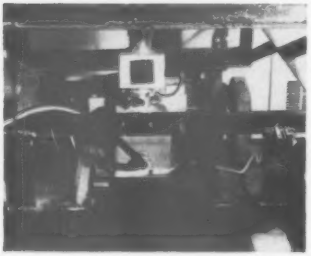

FIG. 3. Vibrating reed and its resistor switchbox, located in base of unit, support weight of ion chamber; this prevents stray currents due to insulator strain. Switch has 6 contacts: 1 for rate of drift, 4 for resistors of 10^7, 10^9, 10^{11}, and 10^{13} ohms, and 1 for shorting electrometer. Dry cells in insulated box behind reed box provide 400-volt collecting current

FIG. 4. Sample is inserted in 16-in.-deep well at top of chamber. Guards and insulation prevent contact with chamber wall which is held at 400 volts. The plastic shielding also serves to limit spread of contamination in the event of a spill

terior of the chamber is exposed when the sample is inserted or removed. The movement due to background radiation was very slight.

Since the efficiency cannot be computed, the minimal detectible activity was estimated as that amount which caused the fiber to move twice as fast as the background rate.

An instrument such as this was used for much of the earlier work on isotopes. The data in the table suggest that the electroscope has potentialities presently being neglected. The thin-window models of the Landsverk or Henson designs could be similarly used, but due to poorer geometry the sensitivity would be less.

End-Window Geiger Counter

A standard, 2.1-mg/cm² mica end-window G-M tube was mounted vertically inside a 2-in.-thick lead shield. The mount for the tube served also as a sample holder; it has a series of slots for inserting the sample holders at varying distances below the window. Because the porcelain cups could not be used with the first slot at 7 mm below the tube, the second slot at 21 mm was taken as the working distance for all samples except those used to show the effect of distance on counting rate and efficiency.

The samples were first counted as 1 ml of liquid and then as an essentially weightless film dried under conditions which ought to minimize or exclude loss of I^{131} by volatilization.

The data in the table show that the count on the dry samples was some three to five times greater than the wet samples at this distance, and that a dry sample in close proximity to the mica window improved the counting efficiency still more. The latter resulted from detection of weak beta particles which had been absorbed by the extra distance, or lost by scattering from the smaller solid angle.

Liquid samples are never placed close to a mica window because the tube becomes unstable; if acidic liquids are counted, the tube life is shortened because of destruction of the mica by acid fumes.

The 1-ml wet preparation in a steel cup was "infinitely thick" and had all the advantages of such sources. The 1-ml liquid preparation in the porcelain capsule gave a higher count because the layer of liquid was only about one-third infinite thickness. But when the sample is less than infinitely thick, small errors in pipetting or imperfections of the cup markedly affect the count. This difference in counting efficiency favoring the porcelain cups disappeared when the samples were dried.

The increased counting rate obtained by drying the sample carries with it two major disadvantages in addition to possible loss by volatilization. One is the difficulty of obtaining an even spread of the dry material; an inordinate amount of effort must be put into preparing comparable samples. The other is that each sample must be corrected for self-absorption according to its net weight. Correction values for this purpose are usually obtained directly by determining the decrease in counting rate as increasing amounts of a representative solid are added to a constant amount of radioisotope. The curves in Fig. 5 illustrate the type of results obtainable.

A simplified theory for self-absorption has been presented (*16*),

$$(N/N_0) = (1/\mu g)(1 - e^{-\mu g})$$

where N_0 is the counting rate without absorber, N is the counting rate with absorber of g grams per cm², and μ is the mass absorption coefficient. The relation between μ and maximum beta-ray energy is given approximately by $\mu = 22/E_{max}^{3/2}$. To obtain the theoretical self-absorption curve for I^{131}, which has 15% 0.32-Mev and 85% 0.60-Mev beta particles, separate curves must be computed for each E_{max} and these then combined with respect to their relative frequencies.

The theory, however, does not describe the increased counting rates when small amounts of solids are present, nor does it take into account the effects apparently related to kind of sample container and counting system, as shown in Fig. 5. Under the circumstances, it seems preferable to correct for self-absorption by curves obtained under conditions identical with those of the data to be corrected.

Thus, counting preparations are set up which duplicate the counting procedures of the experiment in every respect, except that the amount of isotope in each sample cup is constant while the weight of the self-absorbing medium is increased. Each combination of experimental conditions and isotope must have its specific correction curve. The need for such correction can be avoided if all samples, including

standards, can be prepared so that they have the same self-absorption.

The counting rate can be increased by evaporating additional portions of the original liquid sample in the cups, but the solids which accumulate will reduce and finally limit the theoretical increase. Also, this procedure may lead to samples which are not completely homogeneous and so introduce serious errors.

Scintillation Counter

The commercial model used has a NaI(Th) crystal ($1\frac{1}{2} \times 1\frac{1}{2} \times 1$ in.) mounted on the end of a 5819 photomultiplier. Even at the optimal voltage and discriminator settings of the instrument, the background was distinctly higher than that for the G-M tubes. The samples were placed 21 mm from the front face of the crystal so that the sample would be approximately the same distance from the mean depth of the crystal and the sensitive volume of the end-window G-M tube.

The low counting efficiency was due to the poor geometric relation of the sample to the rather small crystal. The effect of distance is shown by a 4.5-fold increase of count when the steel cup was positioned 15 mm nearer the surface of the crystal. The counting efficiency could be improved by using a larger crystal or by devising an arrangement of sample and crystal to give 2π or 4π geometry.

Since the minimal detectible microcurie value was computed in relation to background, the normally high background rate of scintillation counters was largely responsible for the unfavorable figure. The background could be reduced by setting the discriminator at a high level, but the counting rate for I^{131} decreased almost proportionately. However, by counting the samples for longer times so as to get about the same statistics, the net effect would be the detection of a smaller amount of activity.

Since the crystal responded to gamma rays only, little difference was to be expected between liquid and dried preparations of the same sample.

Gas-Flow Proportional Counter

A three-position pre-flush windowless proportional counter was used in conjunction with a "Nucleometer"-type fast linear amplifier and scaler. With methane as the counting gas, a rather good plateau was found at 4,200 volts for the beta particles of I^{131}. Only dry samples were counted. The inner conducting surfaces of the porcelain capsules were carefully grounded to the chamber wall.

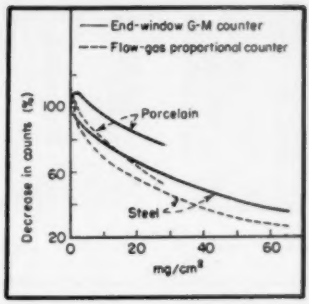

FIG. 5. Self-absorption can cause aberrations in data, requiring calibration of particular apparatus used. As self-absorbing medium (urea here) is added to sample, relation of count to count of weightless sample is not linear. Increase with small amount of absorber is common

The data show that this method of counting was the most efficient and sensitive of those examined. The gain in sensitivity over the common end-window counter was due to the collection and counting of the weak beta particles that never reach the sensitive area of ordinary G-M tubes. Graf *et al.* (*6*) have indicated that such a proportional counter is more sensitive than an analogous system counting in the Geiger region. Another advantage is that high counting rates can be recorded without coincidence losses.

Such sensitivity carries disadvantages with it, however. All except the very weakest samples have to be diluted, and small volumetric errors in preparing dilute samples are magnified in proportion. Similarly, if the samples have slight imperfections, such as areas of variable thickness or density, comparatively large aberrations in counting rate are observed (see Fig. 5).

In practice, therefore, samples in this counter should be either weightless or "infinitely" thick. When it is impossible to obtain weightless or zero-thickness preparations, or when infinite thickness reduces sensitivity too drastically, other more rapid counting methods should be substituted.

The gas-flow proportional counter, according to these tests, provided the greatest sensitivity, but special attention must be given to preparing the samples used in it. Except where the sample must be ashed, the flat-bottomed stainless steel cups are preferable to the treated Coors capsules as counting vessels.

Discussion

The numbers in the table were obtained experimentally with the instruments available and are not presented as a precise evaluation of the best that a particular system can do. In general, as sensitivity of the method increases, the care and time that must be spent on preparing the sample and observing the technical details increase also.

Two of the seven instruments, the "Texas" counter and the high-pressure ionization chamber, are not commercially available. The scintillation counter was representative of the types available commercially at present. The remaining instruments were manufactured by several companies and, as standard instruments, differed chiefly in minor details.

Many organic and mineral samples contain sufficient naturally radioactive K^{40} to be counted by the more sensitive methods. A correction figure can be secured from an I^{131}-free blank, or it can be obtained automatically by adding K ions to standard or reference samples in amounts equivalent to those found in the preparations. KI, KOH, and other potassium salts should be avoided in radioisotope work.

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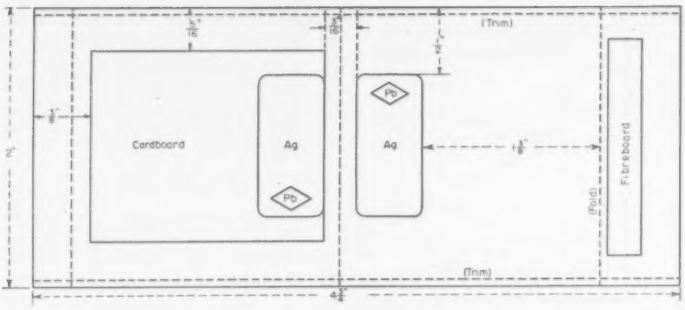
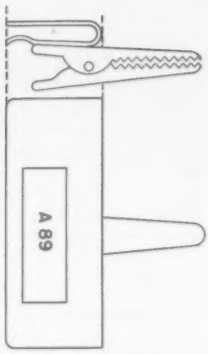


Holder envelope layout before folding. Positioning of the cardboard and fibreboard backing and silver-lead filters is shown



Front and side views of metal closure with alligator clip attached

MATERIALS: for Making Envelope

One piece 2-in. clear polyethylene film tape (No. 480)*—0.005 in. thick, $4\frac{3}{4}$ in. long
One piece 2-in. opaque polyvinyl film tape (No. 471)*—0.005 in. thick, $4\frac{3}{4}$ in. long
Two pieces $\frac{3}{4}$-in. opaque polyvinyl film tape (No. 471)*—0.005 in. thick, $2\frac{1}{4}$ in. long
Two pieces silver sheet—0.020 ± 0.001 in. thick, $\frac{1}{2} \times 1$ in.
Two parallelograms of lead sheet—0.020 ± 0.001 in. thick, 60° angle, $\frac{5}{16}$ in. on a side
One piece 0.027-in. cardboard (file cabinet index dividers)—$1\frac{5}{8} \times 1\frac{3}{4}$ in.
One piece 0.049-in. fibreboard—$\frac{1}{4} \times 1\frac{9}{16}$ in.
Armstrong's industrial adhesive (D236)†—one drop to laminate silver and lead pieces

for Making Closure

One piece soft spring, 0.22-in. sheet brass—$1\frac{1}{2} \times 1\frac{13}{16}$ in.
One piece soft spring, 0.008-in. sheet brass—$1\frac{1}{4} \times \frac{1}{2}$ in. (in which identification number is punched)
One alligator clip (No. 60)
One rivet (AN 453-AD-3-2)

* Minnesota Mining and Manufacturing Co., St. Paul, Minn. † Armstrong Cork Co., Lancaster, Pa.

An Easy-to-Make Film-Badge Holder

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BECAUSE available film badges were unsuitable for our purposes, we developed a film badge employing a silver filter for more accurate evaluation of long-term exposure.† A film-badge holder was also developed having the following advantages over other existing types:

1. Light weight, not much larger than a dental film packet
2. Fairly light-tight and water-tight
3. Built-in shielding for filtration and for determination of the type of radiation
4. Built-in permanent film identification
5. Durability
6. Low unit cost and simple construction

In addition, the plastic holder described here has increased the efficiency of monitoring operations by reducing the man-hours necessary for preparing and processing film badges.

The holder consists of two main parts—the plastic-tape, film-holder envelope, which includes silver-lead-combination filters, and the metal closure on which is punched the X-ray identification numbers and to which is attached an alligator clip. The dimensions of the closed envelop are $1\frac{13}{16}$ in. wide by $1\frac{7}{8}$ in. long by 0.113 in. thick.

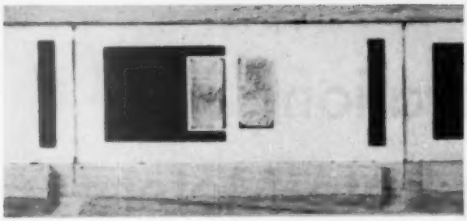
The plastic-film-tape envelope can easily be adapted for wrist or hand monitoring operations by sealing it onto a $\frac{3}{4}$-in.-wide, garter-type elastic wrist band with a strip of clear polyvinyl film tape.

* * *

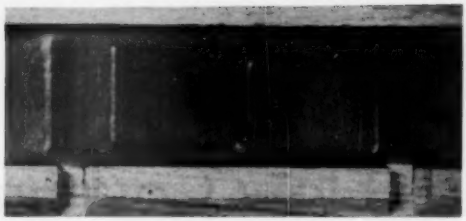
The authors gratefully acknowledge the help and assistance of Mr. Herbert Gass of the Medical Physics Section in the design and construction of various prototype holders before this final design was accomplished. We also wish to thank Dr. Fred A. Bryan and Mr. Reginald Dickinson for suggestions during the course of the development. The work was performed under contract AT-04-1-GEN-12 between the U. S. Atomic Energy Commission and the University of California at Los Angeles.

* Health Physics Department, R-C. Scientific Instrument Co., Playa Del Rey, Calif.

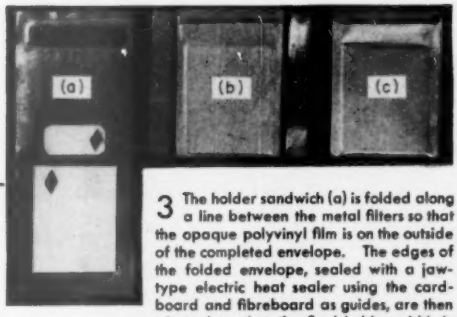
† R. Baker, L. B. Silverman, NUCLEONICS 7, No. 1, 26 (1950).



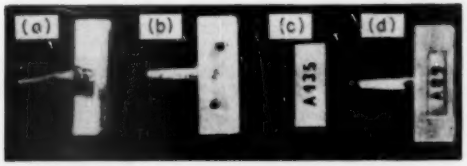
1 Clear polyethylene tape, 16 in. long, is placed adhesive side up in the groove of the wooden template marked off, as indicated in the diagram on p. 62, for assembling three holder envelopes at a time. Laminated silver-lead filters are placed on the tape, over their respective black guide markings, with the lead pieces facing the tape. Cardboard and fibreboard stiffeners for the back and flap are then placed over their guide markings, with the left-hand metal filter covered by the cardboard



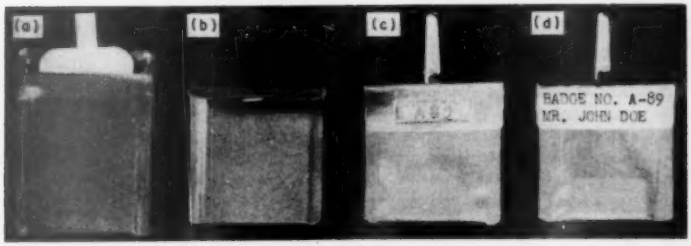
2 Colored, opaque polyvinyl tape, 16 in. long, with the adhesive side facing the clear tape, is pressed onto the polyethylene tape with the metal filters, cardboard, and fibreboard sandwiched between them. Before removal from the template, the three holders are cut apart with a sharp knife at the knife guides provided on the template between neighboring cardboard and fibreboard pieces. The use of a template assures uniformity of badge construction



3 The holder sandwich (a) is folded along a line between the metal filters so that the opaque polyvinyl film is on the outside of the completed envelope. The edges of the folded envelope, sealed with a jaw-type electric heat sealer using the cardboard and fibreboard as guides, are then trimmed so that the final holder width is $1\frac{13}{16}$ in. (b). A $2\frac{1}{4}$-in. piece of $\frac{3}{4}$-in.-wide opaque tape is placed over each sealed edge, so that $\frac{3}{16}$ in. overlaps on each side, and is pressed together (c)



4 The closure, which serves to pinch closed the envelope flap, is cut from 0.022-in. brass sheet as a $1\frac{1}{2} \times 1\frac{13}{16}$-in. rectangle with slightly rounded corners. A window, $1 \times \frac{5}{16}$ in., is cut in the center of and parallel to the $1\frac{13}{16}$-in. dimension, $\frac{1}{8}$ in. from the edge. The rectangle is folded over a $\frac{3}{16}$-in.-radius rod in the form of a "U" clip, with a step in the back, as shown in the diagram on p. 62, to act as a catch on the outside of the fibreboard-reinforced flap (a). An alligator clip is riveted to the back of the closure (b). A piece of 0.008-in. brass, $1\frac{1}{4} \times \frac{1}{2}$ in., with the identification number punched in it (c), is soldered on the inside of the window (d)



5 A $1\frac{1}{2} \times \frac{1}{4}$-in. piece of yellow Scotch tape, folded over one end of a $1\frac{1}{4} \times 1\frac{3}{4}$-in. film packet (shown inserted in the envelope above left) serves as a tab (a). To indicate the film batch number, the packets are arranged front side up on a metal exposure tray, which has a metal cover with $\frac{5}{16}$-in. holes spaced so that the upper left-hand corners of the film packets are exposed. A strip of 0.008-in. brass is placed over the holes with the batch numbers punched to coincide with the holes in the cover. These are exposed to 35-kvp, 10-ma X-rays for $\frac{1}{4}$ sec. The films are placed in the envelope so that the excess tape above the cardboard acts as a light trap and the fibreboard acts as a water trap (b) when the closure is placed on the holder (c). Strips of paper bearing the individual's name and badge number are glued over the brass thin window (d)

6 The film, with the batch number in the upper left corner, is then identified with the badge number by putting the fully loaded badge, with closure in place, in the X-ray unit. The badge is surrounded by $\frac{1}{16}$-in. lead, except for the portion covered by the closure. The number punched in the closure window is exposed to X-rays for $\frac{1}{4}$ sec

Devices for Calibration of Slow-Counting-Rate Meters

For calibration of survey meters, good facsimiles of pulses from the detector are needed in the 0–300 cpm range. Of three units built, one using a repeating timer meets this need with minimum cost, bulk, and weight

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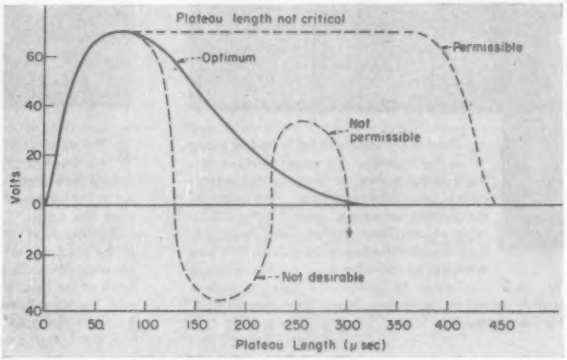

FIG. 1. Pulse shapes for rate-meter calibration

BECAUSE OF THE LACK of commercial equipment and the few convenient pulse sources available, we have developed a unit for calibrating rate meters designed for the relatively slow counting rates of background-radiation monitoring.

Optimum calibration pulses should be an operational facsimile of the output of the G-M tube in use. The general requisite pulse shape, shown in Fig. 1, is a sharp rise to a maximum of about 70 volts, and a smooth decay to zero. A potential reversal terminating the decay is not desirable, since it produces a spurious count in some rate meters. A second rise, produced when the decay is a damped oscillation, is not permissible, since it causes a spurious count in most rate meters. Because calibration pulses are evenly spaced in time, the duration of the plateau at maximum voltage is not critical, but should, in general, be short, and seldom more than half the interval between pulses.

Two Calibrator Types

Before arriving at a calibrator satisfying the conditions of cost, size, weight, and pulse frequency, a unit was designed employing a scaler and a frequency supply. Another design was based on a synchronous motor and a perforated disk. However, scalers and allied equipment would only be available in larger laboratories, and parts for the mechanical system would probably have to be specially made.

Scaler and frequency supply. If a suitable scaler or submultiplier and a convenient supply frequency, such as 60 cycles per sec, are available, it is possible to obtain a pulse output at a low enough frequency. With a series of binary scalers, submultiple outputs of line frequency can be obtained, as in the table on the following page. In the slow-count range, the nonintegral rates are not convenient (for most radiation background

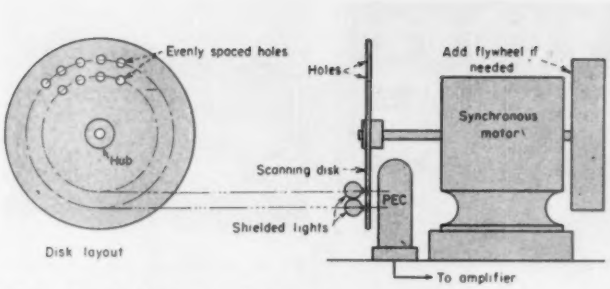

FIG. 2. Photoelectric pulse generator

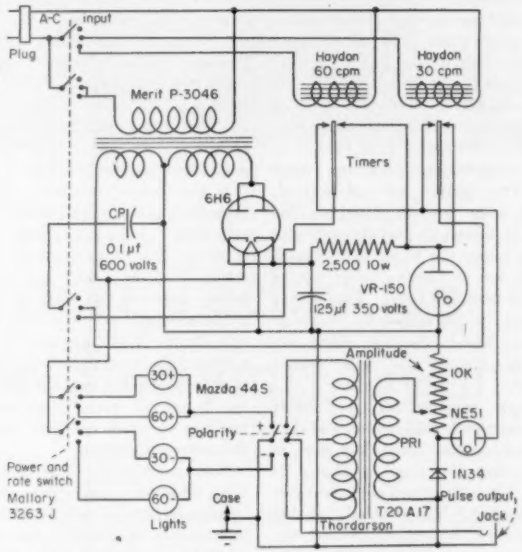

FIG. 3. Circuit of portable calibrator using repeating timer

Pulse Rates Obtained with Scalers and Neon-Ring Submultipliers

Number of stages	*Pulses per min obtained with division per stage by*									
	2	*4*	*6*	*8*	*10*	*12*	*14*	*16*	*18*	*20*
Input = 60 cycles per sec = 3,600 cpm										
1	1,800	900	600	450	360	300	257.1	225	200	180
2	900	225	100	56.2	36	25	18.7	14	10.2	9
3	450	56.25	16.6	7.0	3.6	2.1	1.33	0.875	0.56	0.45
4	225	14.06	2.76	0.875	0.36	useless values				
Input = 400 cycles per sec = 24,000 cpm										
1	12,000	6,000	4,000	3,000	2,400	2,000	1,714.3	1,500	1,333	1,200
2	6,000	1,500	666.6	375	240	166.6	122.4	93.75	74.07	60
3	3,000	375	111.1	46.8	24	13.88	8.74	5.85	4.11	3
4	1,500	93.7	18.51	5.85	2.4	1.15	0.52	useless values		

monitors, two integral values bracketing 40 cpm are desirable). The bulk, weight, and power requirements of a 4–6-stage Eccles-Jordan scaler are also inconvenient.

Another submultiplying method meriting consideration is the neon-ring counter (*1*), which will divide by any even number. The input counts are evenly spaced and of relatively low frequency. Again, as seen in the table, few submultiples are convenient for rate-meter calibration

The best results were achieved with two scalers cascaded. Two frequency values were obtained by using "raw" 60-cycle current and full-wave rectified 60-cycle current. Workable submultiplier combinations are 8 by 10, giving outputs of 45 and 90 cpm; 10 by 10, giving outputs of 36 and 72 cpm; and 12 by 12, giving outputs of 25 and 50 cpm. The two factor-of-12 neon rings were later modified by insertion of switches so that combinations of either 10 by 10 or 12 by 6 could be obtained at will. This, in conjunction with a full-wave rectifier in the input circuit, permits counting rates of 36, 50, 72, and 100 cpm.

A decade Remtron scaler (*2*), replacing the scale-of-10 neon rings, offered no improvement. Both the neon rings and the Remtron are light sensitive, a difficulty that can be eliminated either by using a light shield, or by providing intense, overall illumination. Unless this is done, a bright flash of light in the vicinity of the neon tubes produces a "nervous breakdown" in the scaler. This trouble was particularly severe in one scaler where a strobotron output tube was used.

Under good operating conditions, the accuracy of the output pulses is considerably greater than is usually needed. The number of components subject to failure, however, is quite great. The equipment costs about $200, occupies two cubic feet, and can be moved by one technician of average physique.

Motor and perforated disk. To reduce the bulk, weight, and cost of the calibration equipment, a television technique was tried. A combination of a small synchronous motor, a perforated disk, a photoelectric cell, and an amplifier was used as a pulse generator. Pulse rate, with this device, is determined by multiplying the speed of the motor, in rpm, by the

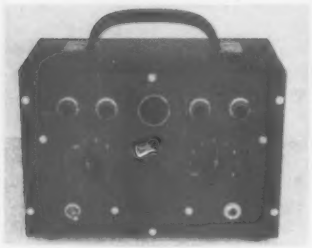

FIG. 4. Front view of calibrator

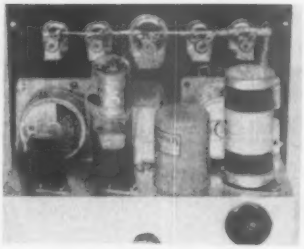

FIG. 5. Interior above-chassis view

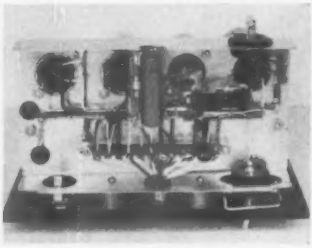

FIG. 6. Under-chassis view of calibrator

number of holes in one circle of the disk. Two rates are provided by drilling two sets of holes on different radii. Switching is accomplished by using two lights, one over each circle of holes. The general arrangement of the components is outlined in Fig. 2. The best combination is a 2-rpm motor and a disk with two circles containing 15 and 25 holes, respectively. This arrangement produces pulses at 30 and 50 per min.

Various interesting, and sometimes useful, pulse shapes can be produced with noncircular disk apertures, or by inserting a shaped mask between the light and the photoelectric cell. Computation of disk and mask-aperture shapes, for a desired pulse shape, may become quite involved.

Although this device has less than half the components of a scaler, costs much less, and is about one third as bulky, it is not suitable for civilian defense use, and can not be made from standard, easily obtainable parts.

Most Satisfactory Calibrator

A standard repeating timer, consisting of an electric clock motor, a cam, and a microswitch, can be used in conjunction with a small regulated power supply, a capacitor, and an output transformer, to produce the requisite pulses at rates within the desired range. Stock 30 and 60-cpm repeating timers, consisting of a 7½-rpm synchronous motor, with either a four or eight-toothed cam, produce the desired pulses with a minimum of auxiliary equipment. A two-rate timer, costing less than $50.00 for parts, can be made in a few hours by any competent technician. The bulk is small, requiring only a standard 5 × 6 × 9-in. utility case, and weight is only 7½ lb.

The counters that we calibrated required pulse output at 30 and 60 per min, of selectable polarity. Pulse height was required to be variable at will, with a maximum of 70 volts, but constant after setting. Pulse shape was not critical, so long as it was an operational facsimile of that produced by a G-M tube, with no damped "trailers" (Fig. 1).

The final circuit is shown in Fig. 3. The clock specifications are rigid as to contacting rate, but all other constants can deviate at least 15% from those shown without impairing operation. The brands are not critical. Special cams can be fitted to the timers, if desired, to provide particular pulsing rates—the timer motors used are capable of turning cams having up to 12 teeth (90 cpm). Two timers are used in preference to a special two-rate timer, because single-cam repeating timers are more readily available.

Chassis construction. A Stanley No. 3 door pull, rubber feet, and 6-32 binding-head case screws are used in constructing the case as shown in Fig. 4. The lights indicate the mode of operation. The control at left energizes the power supply and sets the pulse rate at either 30 or 60. The control at right is for the amplitude potentiometer. The switch at left determines output polarity.

As shown in Fig. 5, a standard steel chassis (Allied 86-040) is used. The electrolytic filter capacitor is mounted as far as possible from the tubes to protect it from heat. The 6H6 tube is the power rectifier; the VR-150 is operated at about one third of its normal current rating. The clock brackets are firmly anchored to the panel to prevent chassis flexure.

The electrical and mechanical construction of this calibrator, shown in Fig. 6, are quite rugged and simple, to facilitate servicing and to prevent in-service failures.

Calibrator operation. Electrical operation of the calibrator is simple and quite straightforward. The power supply produces a regulated output of 150 volts, substantially pure d-c. By means of the timer switch, capacitor *Cp* is charged to 150 volts, then discharged through a neon tube and the output transformer winding. Current through the transformer primary is controlled by the amplitude potentiometer. Flybacks are shorted out by the 1N34 germanium rectifier. Polarity of the output pulses is determined by the output switch.

If the Geiger counters to be calibrated are not equipped with an input jack ahead of the rate meter, a large capacitor (0.1 μf) of adequate voltage rating (such as 1,500) may be connected between the ungrounded side of the calibrator output and the "hot" side of the G-M tube socket. After a 30-sec warm-up period, the calibration pulses are fed into the rate meter for 3 min at each speed, and the readings obtained are compared with the meter calibration.

Four months of use indicates that this rate-meter calibrator performs as planned and as desired. A similar instrument, designed as an integral part of a continuously recording background monitor, is currently inserting calibration signals into the record automatically at regular intervals. The instrument appears suitable for general use in checking G-M rate meters and similar instruments.

* * *

Development of the photoelectric pulse generator was done at Stevens Institute of Technology under the direction of the late Dr. Percy Hodge. Experiments with various scalers were done at Indiana University. Work leading to development of the clock-type calibrator was collateral with the writer's assignments at Cornell Aeronautical Laboratory, Buffalo, N. Y. The gift of a set of Remtron tubes for test purposes, by Remington Rand, Inc., is here gratefully acknowledged.

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FIG. 1. Radioactive gear being placed in gear test machine

FIG. 2. Oil chamber A is directly behind shelf holding scaler

Tracer Technique for Studying Gear Wear

Effectiveness of lubricants can be determined from gear wear data. Wear can be continuously studied by measuring the change in activity of a lubricant circulating through a gear box in which one of the mating gears is radioactive

By V. N. BORSOFF, D. L. COOK, and J. W. OTVOS
Shell Development Company
Emeryville, California

WITH GEARS, as with many other rubbing machine surfaces, lubricants are used to decrease friction, to decrease or eliminate wear, and to cool. These lubricants should be resistant to oxidation or decomposition, should not produce sludges or deposits, and should not attack the parts chemically.

In the process of developing lubricants, all these points are studied. Friction, when possible, is measured directly. The cooling properties of lubricants are evaluated from the temperatures of machine parts and lubricant when running under a specified set of operating conditions. The stability of a lubricant is evaluated from inspection and chemical analyses before and after prolonged service.

Prior Wear Test

A fairly obvious technique for evaluating wear, used extensively in the past, is to weigh the test gear, run it in a test apparatus under specified conditions of load, time, and lubrication, disassemble, and reweigh it to determine the loss in weight. The load-carrying surfaces can also be examined microscopically for visual evidence of wear such as scoring, abrasions, and pitting.

Disadvantages. The disadvantages of this approach are evident. Considerable time is required to produce an easily measurable amount of wear, particularly at low loads. The requirement of stopping the test for each observation, disassembling, cleaning, weighing, and reassembling is also time-consuming; more important, there is always the possibility

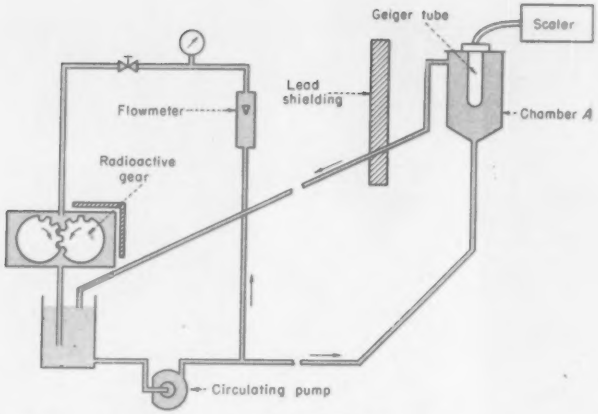


FIG. 3. Gear-wear test apparatus schematically diagrammed

that an extraneous variable (for example, a change in alignment) may be introduced in reassembly. The main disadvantage, however, is that only the total wear between observations can be determined; the initial appearance of wear, variation in rate of wear, and possible transient phenomena escape observation.

Tracer Wear Test

Most of the disadvantages of prior tests are eliminated by the use of tracer techniques. By employing a radioactive gear in combination with a normal gear, and circulating the lubricant through a chamber monitored by a Geiger counter and scaler, a continuous indication of the total amount of metal worn from the radioactive gear can be obtained. There are distinct gains both in speed of test and in continuity and accuracy of observation. The need for disassembly and reassembly, and the consequent danger of invalidating the test because of changed alignment, are eliminated.

The apparatus used for radioactive gear test work is based on the "four-square" principle for loading the gears. It is the same as the unit described in a previous publication* except for modifications in the oil circulating system. The gear box and its apparatus are shown in Fig. 1, the measuring apparatus in Fig. 2, and the general system in Fig. 3.

Part of the oil from the sump is circulated through a chamber, *A*, containing a Geiger tube. The chamber is located at some distance from the shielded test gear compartment. In addition, shielding in the form of lead bricks 2 in. thick is placed between it and the gear box. Chamber *A* is 5 in. in diameter and about 8 in. high with an oil capacity of 1,600 ml; this is about the optimum size for the system, which contains a total of 4,000 ml of oil.

Radioactivity Apparatus

A Tracerlab TGC4 G-M tube, sensitive principally to gamma radiation, is immersed 4½ in. in the oil in chamber *A*. It is used with a model 1000 scaler manufactured by Berkeley Scientific Co. The chamber and scaler are shown in Fig. 2.

Other radiation detecting equipment was also used. A Tracerlab SU-3A laboratory monitor, equipped with a TCG-2 G-M tube and aluminum shield, was used for determinations of the radioactivity of irradiated gears and measurements of metal transferred to the inactive gear. Essentially, this apparatus measures only gamma radiation. The same equipment was used without the aluminum shield in monitoring for cleanliness and stray radiation.

For radiographing the gear surfaces, duPont dental X-ray film, type 52, was used.

To check the intensity of radiation in the laboratory and to establish safe locations for the personnel, a Tracerlab SU-1B portable radiation survey meter was employed. Exposure of personnel to radiation was measured with quartz-fiber electroscopes, model 3, manufactured by Fred C. Henson and Co., Pasadena, Cal. Film badges supplied by the Industrial Health and Hygiene Associates, Berkeley, Cal. were used.

* V. N. Borsoff, J. B. Accinelli, A. G. Cattaneo. The effect of oil viscosity on the power-transmitting capacity of spur gears. *Trans. Am. Soc. Mech. Engrs.* **73**, 687 (1951).

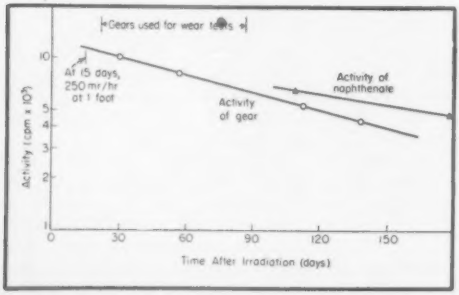

Gears used for wear tests
At 15 days, 250 mr/hr at 1 foot
Activity of naphthenate
Activity of gear
Activity (cpm x 10³)
Time After Irradiation (days)

FIG. 4. Decay curves for radioactive gear and for naphthenate used for calibration

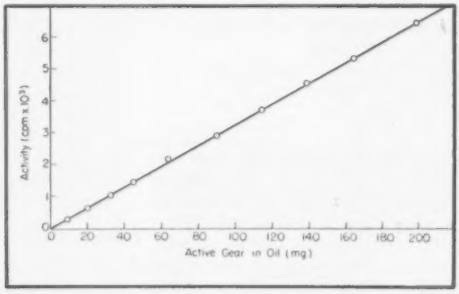


FIG. 5. Calibration curve relating counting rate to weight of gear in oil

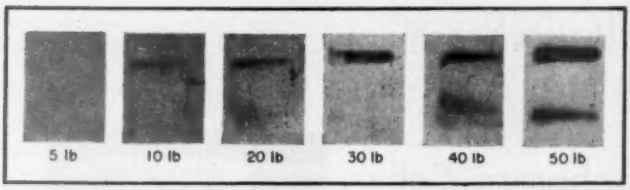


FIG. 6. Radiographs of working teeth surfaces of inert gear after use with radioactive gear at beam loadings shown

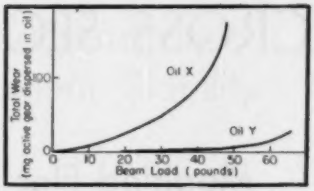


FIG. 7. Typical wear curves obtained by radioactive gear technique

Finally, special tools such as long-handled forceps and special screwdrivers and wrenches, were required for handling the radioactive gears. Visual inspections of the gear-tooth working surfaces were made from a safe location behind lead shielding with the aid of an ASMI Borescope manufactured by the American Cystoscope Co. of New York.

Activity of Gears

Seventeen- and nineteen-tooth spur gears of 6 diametral pitch and 20-degree pressure angle, hardened to 62 RC_c were used in the test gear case. The 17-tooth gears (pinions), weighing about 150 gm each, had been previously irradiated 30 days by neutrons in the reactor at Oak Ridge.

The irradiated gears were periodically monitored under a standardized geometry. The rate of decay thus established is shown in Fig. 4. The average experimental half-life of 90 days agreed well with the calculated value based on analysis of the gears, neutron flux of the pile, and atomic cross-sections for slow-neutron capture. Principal contributors to the activity, discounting a number of isotopes of short half-life whose contributions were negligible after 30 days, were found to be Fe^{55}, Fe^{59}, Cr^{51} and Co^{60}, with half-lives of 4 years, 47 days, 26.5 days, and 5.3 years respectively. The activity of the gears (15 days after removal from the pile) was measured and found to be about 250 mr/hr at 1 ft and between 9 and 10 mr/hr at 5 ft, which is in good agreement with the calculated value of 225 mr/hr at 1 ft for the Fe^{59}, Cr^{51}, and Co^{60}. (The Tracerlab SU-1B used for this measurement would have very low sensitivity for radiation from Fe^{55}).

Calibration

To establish the relation between counts and gear wear, a tooth (2.94 gm) broken from a radioactive gear was dissolved in concentrated hydrochloric acid. Hydrogen peroxide was added to oxidize the ferrous iron to ferric iron, and the solution was evaporated to dryness. The residue was taken up in 100 ml of water, and to it were added 75 ml of carbon tetrachloride and a solution of 165 millimoles of potassium naphthenate (molecular weight of naphthenic acids = 235) in 50 ml of isopropyl alcohol. The ferric naphthenate formed was extracted (by shaking) into the carbon tetrachloride phase, which was separated and used in making up a standard solution of iron naphthenate in oil. Appropriate quantities of this solution were added to the oil in the test apparatus to obtain the calibration curve shown in Fig. 5.

It is to be expected that the cobalt present would appear as cobaltous naphthenate, along with the ferric compound; the chromium might also be converted to a chromic naphthenate. Calibration curves obtained on two different dates indicates a somewhat longer half life (163 days) than for the original gear, as shown in Fig. 4, and therefore would seem to indicate that some of the chromium was not extracted.

Test Procedures, Results

Before each test the irradiated gear was monitored to establish the level of its radioactivity on the day of the experiment. After installing the gears, the test gear compartment was shielded by a lead jacket. Then the radioactivity in chamber *A* was determined, first without the oil, and second with oil flowing. The latter reading established the background for the whole experiment. The machine was then operated under a standardized set of operating conditions and Geiger-counter readings taken at periodic intervals. These readings were then converted to milligrams of wear of the active gear by using the naphthenate calibration. The activity of the gear on the day of test relative to that for the date of the calibration was taken into account.

For studies of metal transfer from the radioactive to the inert gear, the latter was removed, washed, and monitored. It was then placed in a light-proof cabinet with a sheet of X-ray film firmly in contact with the working surface of a tooth. The film, when developed, gave the locations where metal transfer occurred. The relative amount of the metal transferred was evaluated from densities of the films and from the count obtained by monitoring. Typical radiographs are shown in Fig. 6.

Figure 7 is included to show the sort of information which may be obtained by use of the radioactive tracer technique. The beam load, an index of the load on the gears, was increased by small increments (usually 5 lb) at half-hour intervals as the run progressed, and individual points were taken every two minutes during the experiment. The data are plotted in terms of beam load to show more clearly the difference between the two oils tested. At 45 pounds beam load, when the experiment with oil *X* was discontinued, the one with oil *Y* was just beginning to show appreciable wear.

* * *

The authors would like to express their thanks to Drs. V. P. Guinn and C. D. Wagner for assistance in the radioactivity calculations and measurements, to S. S. Sorem and F. C. Younger who carried out the lubrication tests, and to their other colleagues at Shell Development Co. who aided with advice and suggestions.

Correction . . . .

In the article on page 28 and 29 of the September issue, Figures 2 and 3 were interchanged.

Modified Electrophoresis Cell for Determination of Labeled Phosphoproteins

By R. E. HEIN and R. E. CLEGG
Chemistry Department, Kansas State College, Manhattan, Kansas

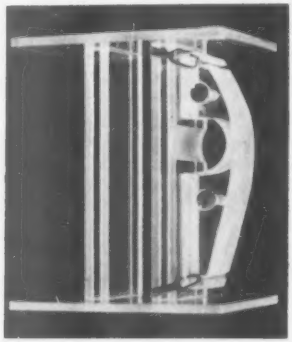


FIG. 1. Modified center cell

A method for observing the amount of labeled phosphorus incorporated in a particular phosphoprotein has been developed. The method consists of a modification of the ascending arm of the center cell of a Tiseleus electrophoretic apparatus so that the radioactivity associated with the labeled phosphoproteins can be measured as the protein fronts migrate past the observation point. The usual information gained from such an experiment as the electrophoretic pattern, protein mobilities, and concentration of the separated protein fractions has not been obviated by this modification.

The maximum energy beta emitted by P^{32} is 1.72 Mev. This means the greatest number of beta particles have an average energy of about 0.9 Mev and only a few greater than 1.6 Mev. A 1.7-Mev beta will be absorbed in 3 millimeters of glass, which is the thickness of the center glass sidewalls of the unmodified electrophoresis cell.

A semicylinder was ground out of the ascending arm two-thirds the distance from the bottom of the cell. A dipping counter tube (1.7 cm o-d Radiation Counter Laboratories, Chicago, Ill.) with a diameter slightly less than the ground semicylinder was held in the desired position by means of a Lucite clamp and rubber bands (Fig. 1). At the thinnest point, 0.9 mm of glass intervenes between the counter tube and serum fractions, thus allowing beta particles of greater than 0.7 Mev to be measured.

The whole assembly is immersed in a water bath at 0.5° C (Fig. 2). Although the leads on the counter tube

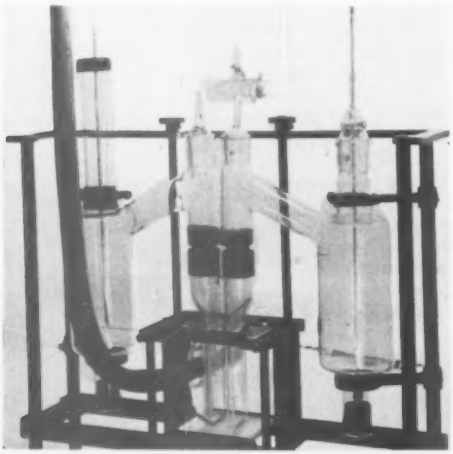


FIG. 2. Electrophoresis apparatus with dipping counter tube in center cell. Apparatus is immersed in a water bath at 0.5° C

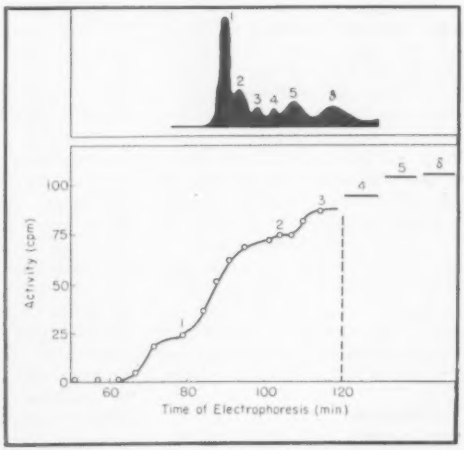


FIG. 3. Top graph shows electrophoretic pattern of 8-week-old chickens. Bottom graph shows associated P^{32} activity

were protected by rubber tubing, moisture condensing inside the tubing caused some trouble until a plug of Drierite was added.

The labeled phosphoproteins were prepared by feeding two 8-week-old chickens radioactive phosphate dried on wheat. The wheat was then fed in gelatin capsules so that the danger of scattered food was minimized. One chicken was fed 0.66 mc/day for three days and the other 0.75 mc/day for four days. At the end of these periods they were sacrificed and serum prepared by centrifugation of the blood clot.

A total of 2.1 μc of P^{32} was present in 8 ml of serum from the three-day-old bird and 2.9 μc of P^{32} in a similar volume from the other. About 0.1% of the fed P^{32} was present in the serum. As indicated by two different experiments, more than 95% of the phosphorus activity in the serum is present as phosphoproteins. In one instance, a sample of the serum was centrifuged in a dialyzing sack with a pore size that differentiates between bound and ionic phosphate.* This indicated less than 5% of the P^{32} existed in the serum as ionic phosphate. In another case after the serum had been prepared for electrophoresis by dialyzing against three portions of sodium borate buffer, about 97% of the activity remained.

A graph of the results from the four-day treated bird along with the electrophoretic pattern is given in Fig. 3. As can be observed, the activity associated with each protein fraction was measured. Once the protein fronts have been separated, it may be advantageous to move the boundaries manually past the counter tube. In the present case, peaks *1*, *2* and *3* migrated by under the electrical field, and *4*, *5* and *6* were moved past manually. The results from the three-day treated bird were similar.

This method, applied to the determination of P^{32} labeled phosphates in phosphoproteins of 8-week-old birds, indicates the greatest activity present in the first two electrophoretic fractions. Similar studies have been carried out with diethylstilbestrol treated birds and will be reported elsewhere.

* * *

The authors are indebted to Mr. E. V. Floyd and Dr. R. H. McFarland for the modification of the electrophoresis cell. The radiophosphorus was obtained from Oak Ridge National Laboratory. This is contribution No. 479 of The Chemistry Department, Kansas State College.

* R. E. Clegg, *Analyst* **38**, 87 (1949).

Natural Airborne Radioactivity as Index of Atmospheric Stability

By RALPH G. SMITH
Bureau of Industrial Hygiene
Detroit Department of Health
Detroit, Michigan

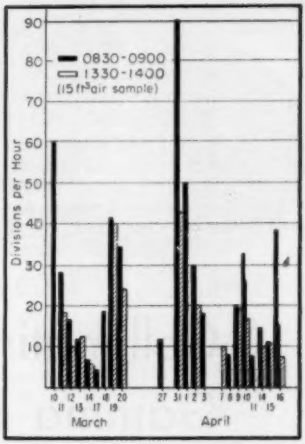
The concentration of a given pollutant in the atmosphere varies with the rate of emission at the source of the pollutant, but the prevailing meteorological conditions can affect the concentration to an even greater extent. Vertical dispersion of airborne materials is as important in this respect as the horizontal distribution caused by wind. Enormous dilutions are caused by vertical mixing and pollution is not important unless present in the lower atmosphere.

Unfortunately, the radiosonde equipment required to measure vertical air motion is very costly. A measurable vector that can be related to vertical stability has been proposed and used by Cowan, however.* He observed that natural background radiation caused by radon and thoron daughter products in the atmosphere could be correlated with temperature differences in the lower 400 ft of the atmosphere.

Cowan's automatic sampling equipment is too costly for the average air pollution or industrial hygiene unit. A simpler method utilizing a quartz-fiber electrometer to measure the activity of filter-paper samples has been devised.

Air is drawn through a 1-in. disk of filter paper at the rate of 0.5 ft³/min for 30 min, resulting in a 15-ft³ sample. The disk is placed in the chamber of the electrometer and an initial reading is taken within 5 min. A second reading is taken 60 min later. No attempt was made to convert these readings to absolute radiation units; instead, the difference between the two scale readings is used as an index.

This procedure was carried out over a two-month period with two samples a day taken at about the same time each morning and afternoon. Samples were not collected on week ends and holidays. The results are shown in the illustration. The index ranges from about 2 to 90 scale divisions/hr. Readings taken at closer intervals showed that the atmospheric radioactivity had a half-life of about 40 min during the first hour of measurement.



DAILY SAMPLES of filter paper used to collect atmospheric dust exhibited this radioactivity. Correlation is possible with atmospheric stability

Good agreement was noted between the index and the prevailing weather. An artificial suppression of activity seemed to result when snow on the ground hindered release of the daughter products. An increase in activity was noted for periods of rainfall; this is caused by washing down of radioactive dust from the upper atmosphere.

From the limited data collected, it cannot be said that the index provides an infallible measure of atmospheric stability. The index should be useful, however, as an aid to comparing air samples collected under varying meteorological conditions.

* * *

This is a condensation of a paper presented before the American Industrial Hygiene Association, April 24, 1952, Cincinnati, Ohio.

* F. P. Cowan, "Quantitative summary of natural radiation and naturally occurring isotopes," lecture presented at the In-service Training Course in Radiological Health, School of Public Health, University of Michigan, Ann Arbor, Mich., 1951.

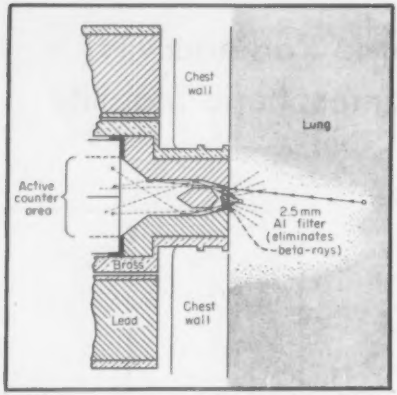


FIG. 1. Cross section of collimating device for measuring gamma rays selectively

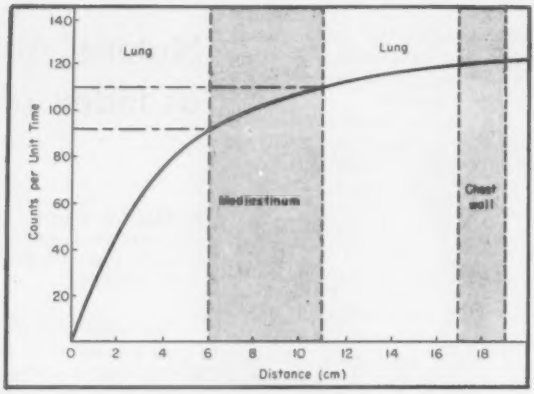


FIG. 2. Effect of extrapulmonary radioactivity. A 30% change in chest wall and mediastinum causes only 4.8% change in total count

Collimating Device for Measuring Gamma Rays from a Limited Region

By ALLEN F. REID and BEN WILSON
Departments of Biophysics and Surgery
Southwestern Medical School, University of Texas, Dallas

For some purposes it is important to limit the measurement of radioactivity primarily to gamma rays from a certain region. This may be accomplished by blocking off the region with lead shields. In certain cases, however, this shielding cannot be done, such as when measuring the amount of I^{131} in the lung of a living dog while excluding most of the radiation from I^{131} in the rest of the dog.* This problem stimulated the development of a collimating apparatus and technique that can be used to measure gamma rays from a near source with minor interference from an unshielded gamma-ray source at a greater distance in the same direction from the detector.

The method uses heavy shielding to admit selectively gamma rays from a definite region into the collimator. None of these gamma rays enter the detector proper, but a certain portion of them is absorbed on the inner surface of the collimator giving secondary radiation which is measured. This can be seen in Fig. 1, which diagrams a typical arrangement in which the gamma-ray activity of a portion of a lung is to be measured. A portion of the collimator is inserted through the chest wall and against the lung. A considerable fraction of the gamma rays from directly in front of the lumen of the collimator is absorbed on the collimator wall at positions from which secondary radiation may proceed to the "active counter area." At greater distances in this direction, however, smaller and smaller fractions of the gamma rays can reach the effective area inside the collimator. Combined with the inverse-square and mass-absorption effects, this collimation delimits counting to the lung.

The effectiveness of regional limitation using the collimator is illustrated by Fig. 2. Counts were taken of increasing thicknesses of an I^{131} solution placed in front of the collimator; these measurements showed there was comparatively little effect on the total count from the more distant layers. Representative activity values could thus be determined for the lung, realizing that variations in activity in other structures would cause a relatively slight change in the measurements.

It is evident that for optimum results the inside of the collimator tube should be lined with material of high atomic number so as to have a high output of secondary rays from the surface. The detector used should have a low sensitivity to gamma rays and a relatively high sensitivity to the secondary rays, thus minimizing the effects of the gamma rays penetrat-

* B. Wilson, A. F. Reid. The effects of hemorrhage and intravenous and intra-arterial infusion upon respiration and the volume of blood in the lungs, "Surgical Forum," Clinical Congress of the American College of Surgeons, pp. 12-18 (W. B. Saunders Co., Philadelphia, Pa., 1951)

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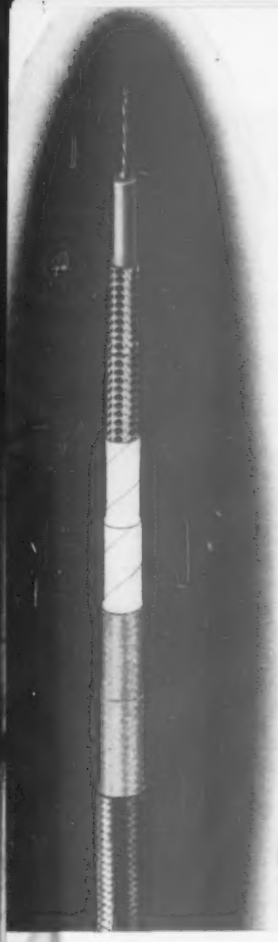

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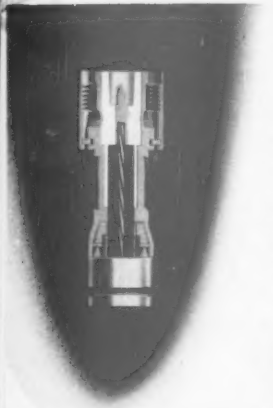

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ing the shielding. A further refinement may be obtained by making measurements with a filter inserted immediately in front of the detector to cut off the secondary rays. The difference between the measurements with and without the filter then serve as quantitative determinations of the radioactivity of the region.

Several modifications of the design shown have been successfully used for various experiments where the basic problem was the same. The device is of little value except for those instances in which it is necessary to minimize the variations in count from unmeasured changes in radioactivity in the area contiguous to the structure under study.

TECHNICAL BRIEFS

- **Wetness of soils** is determined by measuring scattering back of neutrons from the soil. Radium-beryllium source of neutrons is used; activation of indium foil is proportional to degree of wetness. The method, developed by J. W. T. Spinks at University of Saskatchewan, Canada, is reported in *Canadian Isotope Newsletter*.

- **Sintered aluminum** has greater fatigue strength at high temperatures than aluminum that is normally processed. Powder-metallurgy technique developed by A. von Zeerlader of Neuhausen, Switzerland, produces aluminum with tensile strength of 50,000 lb/in² at 300–400° C.

- **Lack** of high-purity material has made metallurgical work with rare elements such as zirconium almost impossible in Europe, according to Dr. H. Hausner of Sylvania Electric Products, Inc.

- **Electrical activity** of atmosphere is measured with aid of radioactive point on top of slender shaft. R. E. Falconer, General Electric meteorologist, has used the device to indicate cold and warm fronts, thunderstorms, and to count lightning strokes.

- **Silver plating,** used to minimize attenuation loses in microwave components, is activated with neutrons from a radium-beryllium source in a thickness test reported by W. H. Colner and H. T. Francis of Armour Research Foundation. Preliminary investigations indicate that the intensity of the induced radioactivity is a purely linear function of plating thickness.

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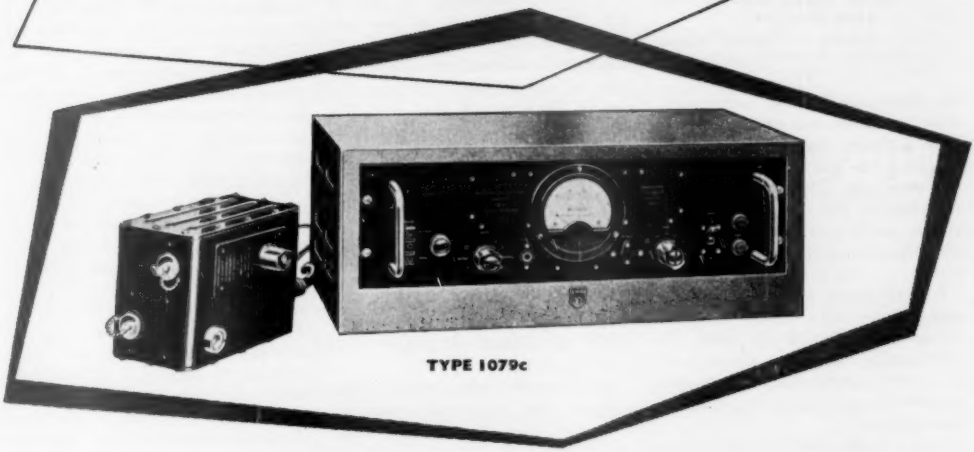

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BOOKS

Atomic Power—An Economic and Social Analysis

By WALTER ISARD and VINCENT WHITNEY (The Blakiston Company, New York, 1952, xi + 235 pages, $4.75)

Reviewed by JOHN R. MENKE, Nuclear Development Associates, Inc., White Plains, N. Y.

Drs. Isard and Whitney have undertaken to answer just about the same large questions that Drs. Schurr and Marschak* so ably posed and studied two years ago. They have examined the probable costs of electricity via atomic energy and have investigated the effects, economic *and social*, of these costs.

In considering social and cultural aspects, they have usefully extended the scope of the previous work. In re-examining the ground already covered, they have exercised much critical judgment; in some cases, they are very critical indeed.

It is not altogether the fault of the authors that the first two chapters dealing with the technical background and the costs of nuclear reactors are lengthy and discursive. For many of the fields have not "boiled down" enough for succinct expression. (There is not yet even a good technical book on reactors.) And for all of us, the areas of ignorance about costs are still large. Yet the authors indicate in Chapter 1 the directions for simplifying the discussion. On page 19 they reach an important and correct conclusion, "But with breeding . . . the likelihood of resource exhaustion is removed as a barrier to atomic power production." Then why the long discussions of nonbreeders? On the same page, speaking of uranium *and thorium* supplies, they indicate a theoretical possible factor of 140 for multiplying the power extracted from minable reserves by breeding. This maximum factor of 140 applies to uranium *alone;* for uranium *and thorium* it is probably nearer to 700.

On page 22, a minor (?) slip cuts the atomic expansion plans from billions to millions.

In Chapter 2, all of the cost estimates already given in the literature are reviewed. It seems that the authors chose to take the least optimistic estimates as being most nearly correct because of the lack of actual economic accomplishments to date. Your reviewer questions the logic of this choice. He believes that the breeding reactors with at least two major products to sell, power and fissile material, will demonstrate the correctness of the more optimistic cost estimates even with a *non*subsidized price for the fissile product. In considering the capital cost goal for nuclear reactors, the authors have seemingly given no credit to this second product.

It is disappointing that the authors did not expand on a major economic problem of the breeder reactor, the "insoluble theoretical problem . . . of allocating the costs of a production process to multiple products," mentioned on page 27.

In the conclusion to Chapter 2, the authors avoid (even more than did Schurr and Marschak) taking a stand on what the costs of atomic power will be. They indicate that such a question is now "too speculative to be subjected to objective economic analysis." But this is just the question that must be faced squarely by the various groups of enterprises now studying the field of atomic power.

Happily, the authors shed extreme pessimism in the concluding Chapter 11 and give a balanced prospect for a useful, perhaps not revolutionary, but sound and economical atomic-power industry. This prospect is not logically derived but is, and perhaps properly so, a matter of faith.

The larger part of the book, Chapters 3 through 10, is devoted to a critical examination of the probable economic and social effects of assumed atomic power. It is an incisive and sensitive job and multiplies manifold the usefulness of the earlier investigations of Schurr and Marschak. The authors have examined very carefully the "other things being equal" clauses and have shown that in important respects other things never are.

In bringing up the cultural obstacles to technical innovation, they have performed a large service. One hopes they will continue and extend their discussion to include a study of these obstacles in the situation on the North American continent.

In the location and industrial economic studies, the authors have taken a very worth-while and objective view of atomic effects. They have shown the innovation to be perhaps significant, but not revolutionary or disruptive. (A favorite example of this nondisruptive effect is to show the percentage of a utility's total investment that might be affected by atomic power replacing *all* of the ordinary fuel-fired boilers. It is approximately 17%*—and that percentage is for the industry most affected!) Atomic power is perhaps not the world-changing and spectacular force as sometimes advertized; it is, however, one more potentially important industry, not at all negligible for intelligent planning.

In discussing specific industries (and particularly iron and steel, and aluminum), the authors indicate how many other important considerations besides low-cost power go into a determination of location. They discuss the probable effects of atomic power on economic development in more and less industrialized economies. As an indication of the conclusions reached, they have made a very good case for the thesis that atomic power will have most immediate application and utility in the *more* industrialized areas rather than in those that are just starting to industrialize their economies. The high cost of capital, and the nonavailability of skills and technologies are some of the factors mitigating against the growth of this new power source in regions most in need of more power.

An interesting and detailed examination of Brazil as an industralizing economy is given, indicating all the specific difficulties to the introduction and application of atomic-energy plants. Also indicated is the large number of other factors required to increase the productivity and standard of living in Brazil; among these requirements, atomic power does not stand out as a single major factor. Rather its effect will be found indirectly as of one strand lifting a complicated warp and woof.

This book is a stimulating and worthwhile work, understandably controversial in the Immediate discussion of atomic costs, but probing, sensitive, and imaginative in the examination of atomic effects on the world. It should be included in the libraries of the nuclear and economic fraternity, and

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* S. Schurr, J. Marschak, "Economic Aspects of Atomic Power" (Princeton University Press, Princeton, N. J., 1950)

* Figure given by W. L. Cisler, president of Detroit Edison Company, in talk on electric power systems and nuclear power, Fifth Annual Summer Institute, University of Michigan Law School, Ann Arbor, Mich., June, 1952

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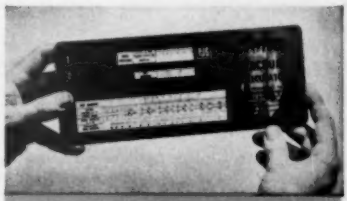

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the work itself should be continued and extended. For, "If there are final answers, they lie in the future."

BOOKS RECEIVED

High-Energy Particles, by Bruno Rossi, Prentice-Hall, Inc., New York, 1952, xii + 569 pages, $12.50. (*To be reviewed.*)

Theoretical Nuclear Physics, by John M. Blatt and Victor F. Weisskopf, John Wiley & Sons, Inc., New York, 1952, xiv + 864 pages, $12.50. (*To be reviewed.*)

Physical Foundations of Radiology, 2d ed. (revised and enlarged), by Otto Glasser, Edith H. Quimby, Lauriston S. Taylor, and J. L. Weatherwax, Paul B. Hoeber, Inc., New York, 1952, ix + 581 pages, $6.50. (*To be reviewed.*)

Advances in Electronics, Vol. IV, edited by L. Marton, Academic Press Inc., New York, 1952, x + 344 pages, $7.80.

Like its predecessors in the series, this volume is a quality presentation of reviews of new developments in particular electronic systems. Seven reviews are offered.

Of particular interest is a review on *The Scintillation Counter,* by G. A. Morton (Radio Corporation of America). He covers the photomultiplier, multiplier performance, phosphor crystals, and scintillation counter applications (23 references). Another review, *Electron Scattering in Solids* by H. S. W. Massey (University College, London), discusses elastic, inelastic, and multiple scattering, energy loss, and mobility of electrons in metals, alloys, and semi-conductors.

The Auger Effect and Other Radiationless Transitions, by E. H. S. Burhop, Cambridge University Press, New York, 1952, xiv + 188 pages, $5.50.

The study of artificial radioactivity and of the possibility of producing inner-shell ionization by *K*-capture processes and the internal conversion of gamma radiation has brought about renewed interest in radiationless transition processes of the Auger type, associated in the past only with the interpretation of X-ray spectra. This monograph is a discussion of the Auger effect that is basically descriptive and experimental. In connection with its relation to internal conversion of gamma-rays, important work in the

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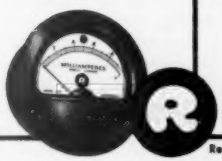

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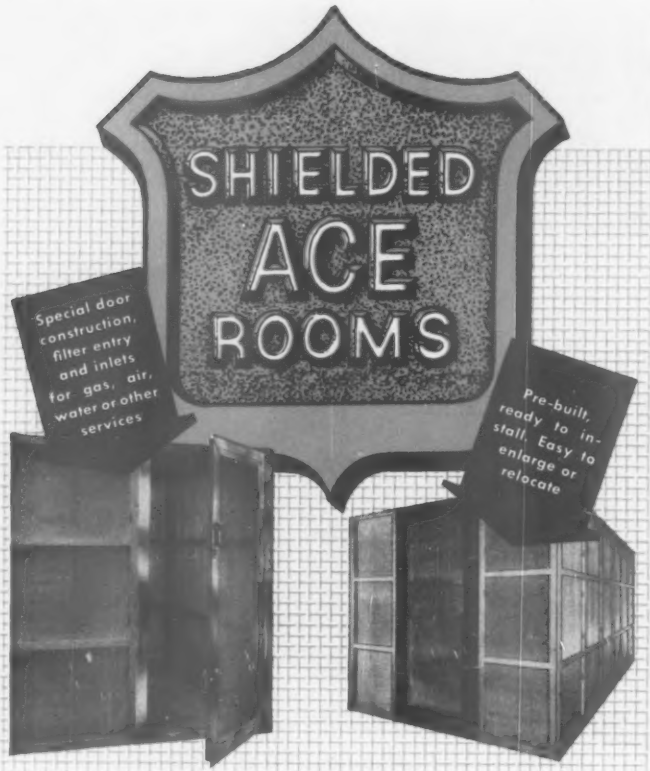
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field up to December, 1950, is covered. Data are included on internal conversion coefficients and fluorescence yields. A chapter is devoted to radiationless transitions in atomic and molecular spectra, and another to the Auger effect in meson capture.

Isotopes in Biochemistry, edited by G. E. Wolstenholme *et al.*, The Blakiston Company, New York, 1952, 288 pages, $5.

Proceedings of a symposium arranged by the Ciba Foundation and held in London last year.

Dictionary of Civil Defence, edited by Carlton Wallace, The Philosophical Library, New York, 1952, 160 pages, $2.75.

Information, given in encyclopedic tabulation, on all phases of civil defense as practiced in Great Britain where community preparedness is actively organized.

Dictionary of Conformal Representations, by H. Kober, Dover Publications, Inc., New York, 1952, xvi + 208, $3.95.

This book is a listing of conformal mappings classified according to the analytic functions describing the transformations. It is offered as an aid in solving the Laplace equation in two dimensions for many boundary conditions in problems such as those related to electrostatics, magnetic fields, heat flow, gas dynamics, hydrodynamics, etc. The collection was made for the British Admiralty in 1944–1948.

Cosmology, by H. Bondi, Cambridge University Press, New York, 1952, 179 pages, $4.50.

The author, disclaiming cosmology as a minor branch of general relativity or as a branch of philosophy and logic, regards it rather as a branch of physics in its own right. His aim in writing this contribution to the Cambridge Monographs on Physics is to say so. He discusses principles, observational evidence, and theories. The latter section, the longest part of the book, includes Newtonian and relativistic cosmology, kinematic relativity, the steady-state theory, and the theories of Eddington, Dirac, and Jordan.

Ultraviolet Radiation, by Lewis R. Koller, John Wiley & Sons, Inc., New York, 1952, ix + 270 pages, $6.50.

This is a broad-interest book, with a wealth of technical information compiled to answer the questions of

the nonspecialist who must use radiation at the short-wave end of the spectrum. Medical men, chemists, engineers, physicists, biologists, and interested laymen, take note.

OTHER LITERATURE

Major Activities in the Atomic Energy Programs (January–June, 1952). A reprint of the Twelfth Semiannual Report of the U. S. Atomic Energy Commission to Congress. *GPO, Washington 25, D. C., $0.35.*

A Glossary of Uranium- and Thorium-Bearing Minerals, by Judith Weiss Frondell and Michael Fleischer. A second and revised edition of Geological Survey Circular 194. It contains 350 entries comprising 182 species of minerals. *Director, Geological Survey, Washington 25, D. C., no charge.*

Research in Engineering Economics. A free-for-the-asking pamphlet on the ways in which Battelle Memorial Institute's service in this field can aid management in industrial planning. Nucleonics is among its research activities. *Battelle Memorial Institute, 505 King Ave., Columbus 1, O.*

Note-book Size Periodic Charts. A handy, two-color, three-punch, 8½ × 11 inch chart of the elements, with logarithms and electromotive force tables on its reverse side. *Fisher Scientific Company, Pittsburgh, Pa., $4.50/pkg of 100.*

Tables of Coulomb Wave Functions, Vol. I (National Bureau of Standards Applied Mathematics Series 17). NBS has published this 141-page volume to stimulate interest in this branch of physical investigation. It contains tabulations of the regular solutions of an important differential equation which arises in the quantum mechanical treatment of two particles moving in a Coulomb field of force, particularly in problems of *p-p* and *p-n* interaction. (Previous tabulations have been comparatively incomplete in respect to both range of parameters covered and size of interval.) A foreword by G. Breit of Yale University and an introduction by M. Abramowitz of the NBS Applied Mathematics Laboratories offer a discussion of the general position and application of the Coulomb function in nuclear physics. *GPO, Washington 25, D. C., $2.*

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NUCLEONIC EVENTS

AEC Sees Ceiling on Need for A-Bombs, Plans to Develop Peaceful Uses of Atomic Energy

With its multibillion dollar expansion program under way, the Atomic Energy Commission now plans to push the peaceful uses of atomic energy. To facilitate this, the AEC is actively considering relinquishing part of its monopoly in this field.

In the first statement of its kind made by a commissioner, AEC Chairman Gordon Dean said in a talk in San Francisco last month that there are two reasons for this attitude: (1) there is a ceiling on the number of atomic bombs—and thus fissionable material—which the military needs, and (2) prospects are now the brightest they have yet been for peaceful applications of atomic energy.

Dean pointed out that there is a law of diminishing returns which predicts that in the forseeable future we will reach the point where we have all of the atomic weapons we can use. In anticipation of reaching this point, Dean feels that we must make a determined effort "now" to develop peaceful uses of atomic energy—because "it is the peaceful promise of atomic energy that has captured the hope and imagination of the peoples of the world" and because we shouldn't let moss grow on our billion dollar plants after they have served their military purpose.

Prospects for peaceful uses of atomic energy look bright, according to Dean, because of two things: (1) of the four industry groups studying the economic feasibility of nuclear power plants, not one has made a pessimistic report to AEC, and (2) AEC technological progress has resulted in downward revision of cost estimates for nuclear power plants.

As an example of cost reduction, Dean mentioned a recently completed reactor (presumably the Materials Testing Reactor in Idaho) which cost $20-million but which, with minor modifications, might now be built for $5-million or less.

AEC is "very seriously looking into" the possibilities of a reactor of this cost, according to Dean, which might be "competitive in a few isolated locations where normal power costs are extremely high . . . What I am talking about here is a relatively small machine that would produce kilowatts numbered in the thousands but sufficient to perform a useful purpose in undeveloped or underdeveloped areas which are remote from normal sources of power, and which are now either priced out of the powermarket or can afford only a marginal supply at best."

With the AEC having emphasized military applications in the past, Dean says that he and his fellow commissioners feel that peaceful uses should be developed now and that "our atomic enterprise and country is big enough to accomodate both a program for defense and a program for peace."

But if peaceful uses are to be pressed, AEC feels that the Government must "relinquish at least part of its monopoly."

Building of 20,000-kw Nuclear Power Plant Would Open Up Mining and Irrigation Projects

A "crying economic need" by the mining industry of western United States for lower-cost power could be met today by nuclear power plants. Stating that the public has paid for the development of nuclear power and that "delivery should not be delayed," U. S. Senator George W. Malone of Nevada last month called for the Atomic Energy Commission to build a 20,000-kilowatt nuclear power plant near Eureka, Nevada (NU, Sept. '52, p. 66).

"Many mining districts throughout the west have long been dormant," according to Sen. Malone, "only because the distances are too great to transmit the relatively small amounts of power for which they can guarantee payment before development.

"In Eureka, where, it is conservatively estimated, there is located at least $100-million worth of zinc and

ACTIVATION ANALYSIS OF MATERIALS requires the sample to be so active that it must be handled behind a lead shield. The sample shown being removed from a lead container has been irradiated in the Oak Ridge graphite reactor. Measurement of the induced activities will tell what impurities are present. (For description of the method, see NU, Mar. '51, p. 62.) This analysis technique is now being offered as a service to industrial, scientific, and medical organizations. For information, write to Radioisotope Control Department, Oak Ridge National Laboratory, Box P, Oak Ridge, Tennessee

lead in one mine alone, and from which it is estimated that 12,000 gal/min of water must be pumped from a depth of 2,500 ft to make such values available—and where the diesel power costs 3 cents per kwh—the proposed 20,000-kw nuclear power plant would save millions of dollars in the production of these strategic metals, and effect a great saving in the cost of plutonium production for the Government.

"It will make both mining and irrigation projects feasible. For example, the power users could pay the Government 1¼ cents per kwh (estimated cost of nuclear power), money now lost to the Government, and the power users would reduce their costs 1¾ cents per kwh.

"Within a radius of 75 miles of Eureka, several small electric utility companies now using diesel power would welcome interconnections with nuclear power. Within the same radius, several large mining companies, such as Gold Acres and Simplot Iron, are ready to contract for power; and a large number of ranchers want power as soon as possible for pumping groundwater. Thousands of acres in eight large desert valleys nearby only need reasonably priced power to be reclaimed.

"For a time the Eureka district hoped it might obtain power from Boulder Dam, some 300 miles south, but the distance was considered too great for economic transmission to supply existing needs. In general, no hydropower is available there.

"Commercial nuclear power is feasible in the high power cost mountain and desert areas now . . . All great inventions that changed the course of history were crude at first . . . but every time a plant is built something new will be learned to reduce the cost."

AEC Technical Staff to Grow by 4,500 in 4 Years

An estimated 4,500 scientists and engineers, in addition to replacements for people currently employed by the Atomic Energy Commission, will be required over the next four years, according to Commissioner T. Keith Glennan, in an address to the American Society for Engineering Education.

To indicate the level of education required of these men, Commissioner

The Editors Hear . . . .

• The production of uranium-235 and plutonium-239 is now big business. "Some day, even in peacetime, the nucleonics industry will be bigger than the iron and steel industry" is the opinion of Zay Jeffries, General Electric vice president.

• Homogeneous reactors may hold the key to the successful development of a nuclear power industry. One analysis shows that, with homogeneous reactors, the cost of generating nuclear power will be very significantly lower than the cost of conventional power in remote areas of the U. S.—and it might even be competitive in nonremote areas.

• Wide use of thorium for the production of fissionable uranium-233 is awaiting the successful development of the breeder reactor. Canada is producing U-233 in small quantities and is now operating a plant for separating it from irradiated thorium.

• Disposal of radioactive wastes at sea has been done to a limited extent by Government agencies. Now five industrial concerns—two in Boston, one in New York, one in Berkeley, and one in New Orleans—have been approved by the AEC for disposal of wastes at sea.

• Industry in New England has been advised by A. D. Little, Inc., to explore the possibility of nuclear power's being the answer to the region's high power costs.

• Sixty per cent of the work at Harwell, Britain's atomic energy research center, is on peaceful applications of atomic energy.

• It is not now clear whether a natural uranium dual-purpose reactor (for the production of plutonium and power, with power as the primary product) could produce as much plutonium as the Hanford production reactors for the same investment of fuel.

• The long-term role of our tremendous gaseous diffusion plants is a question mark. If there is no military need for their use in the production of enriched uranium, they will undoubtedly be too costly to be considered as part of an industrial nuclear power system . . . . With these plants in use, dual-purpose reactors will need only enough enriched uranium to start up; thereafter they can depend upon the "leavings" of the diffusion plants—the unenriched uranium which could be converted into fissionable material.

• Location of power reactors with respect to public safety is a controversial subject these days. General sentiment in industry is that stringent AEC requirements will have to be relaxed. Regulations are now set up based on maximum possible hazards which can develop, e.g., the exploding of a reactor.

• Although a lot of useful technical data are being obtained in work on military reactors, much of the designs would be useless for industrial reactors because of the special requirements of the military, particularly the Navy.

• Upwards of a dozen universities are now interested in the possibilities of obtaining research reactors for campus use. AEC has delayed announcing any policy on this.

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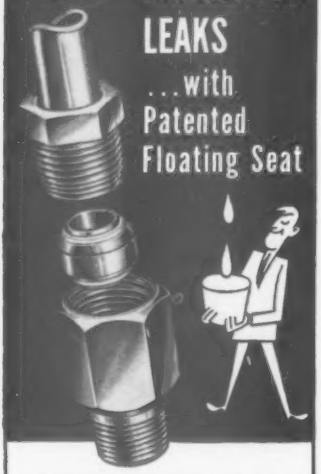

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Brush Beryllium Co., Luckey, Ohio	375
Carbide and Carbon Co. (C-31), Paducah, Ky.	1,148
Carbide and Carbon Co. (Y-12), Oak Ridge, Tenn.	4,298
Carbide and Carbon Co. (ORNL), Oak Ridge, Tenn.	2,576
Carbide and Carbon Co. (K-25), Oak Ridge, Tenn.	5,574
Dow Chemical Co., Boulder, Colo.	369
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General Electric Co. (Services), Richland, Wash.	534
Iowa State College, Ames, Iowa	558
Mallinckrodt Chemical Co., St. Louis, Mo.	704
Management Services, Inc., Oak Ridge, Tenn.	1,319
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Westinghouse Electric Corp., Pittsburgh, Pa.	1,366
Zia Co., Los Alamos, N. M.	1,466
All other operations contractors	5,652
AEC Offices	
Chicago	315
Hanford	452
Idaho	444
New York	637
Oak Ridge	1,337
Raw Materials (Field)	248
San Francisco	77
Santa Fe	1,689
Savannah River	329
Schenectady	63
Washington	1,146
Construction Contractors	
Atkinson and Jones, Richland, Wash.	4,031
Atkinson and Jones (Minor), Richland, Wash.	705
Austin Co., Denver, Colo.	1,770
Bechtel Corp., Arco, Idaho	321
California Research and Development, Livermore, Calif.	1,351
Catalytic Construction Co., Philadelphia, Pa.	635
DuPont, Augusta, Ga.	35,880
Fuller Construction Co., Cincinnati, Ohio	3,686
Girdler Corp., Newport, Ind.	2,024
Holmes and Narver, Los Angeles, Calif.	1,539
F. H. McGraw and Co., Paducah, Ky.	21,908
Maxon Construction Co., Oak Ridge, Tenn.	896
R. E. McKee, Los Alamos, N. M.	658
Silas Mason Co., Amarillo, Texas	453
Utah-Leavell, Los Alamos, N. M.	365
All other construction contractors	4,756

Glennan cited the following figures. Between July 1, 1952, and June 30, 1953, Oak Ridge National Laboratory will hire 30 scientists with doctor's degrees, 45 with master's degrees, and 95 with bachelor's degrees.

Brookhaven National Laboratory has been employing about 40 men with doctor's degrees and 20 with master's or bachelor's degrees each year. Since the major projects at Brookhaven—the reactor and the cosmotron—are completed, or nearly so, this number will be down to about 30 qualified technical men each year.

The staff at Argonne National Laboratory is fairly well stabilized. Last year 26 Ph.D.'s, 17 Masters, and 26 Bachelors were hired to replace 61 persons who left.

At Los Alamos, the estimated hiring for the next year will be 66 Ph.D.'s, 30 Masters, and 74 Bachelors.

The breakdown of the present staff at Los Alamos is 266 Ph.D.'s, 121 Masters, and 287 Bachelors.

The table above shows the total number of persons employed by AEC.

AEC Accepts New Nuclear Power Study Proposal

The Atomic Energy Commission has accepted a proposal by the Pioneer Service and Engineering Co., Chicago, and the Foster Wheeler Corp., New

EXPERIMENTAL NUCLEONICS

By Ernst Bleuler and George Jason Goldsmith, both of Purdue University
393 pp. $6.50

Primarily a handbook for students in radioactive tracer technology, nuclear engineering, and nuclear physics, this book includes experimental procedures related to the measurement of radiations from radioactive sources, as well as information in regard to setting up and operating a tracerlab. Copiously illustrated with numerous tables, diagrams, and graphs, the book includes the Segre chart of the isotope.

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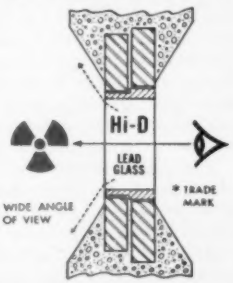

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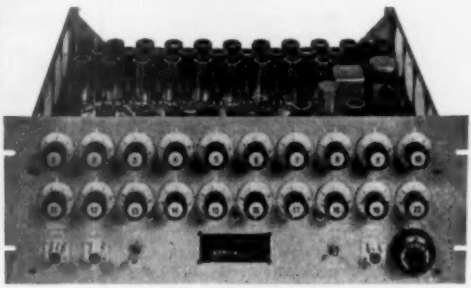

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York, to conduct a 12-month survey of the feasibility of design, construction, and operation by private industry of power-producing reactors.

The proposal is for a study similar to those which were conducted last year by four groups of firms (NU, June '51, p. 42, July '51, p. 2). Any agreement for this new study, or for any other for which a proposal is submitted, will follow the same lines as the first round of contracts. The main goals of the studies include:

1. Determination of engineering feasibility of design, construction, and operation of a reactor by private enterprise.

2. Examination of economic and technical aspects of building such a reactor.

3. Measure of research and development work needed before such a reactor project could be undertaken.

4. Recommendation to AEC as to what should be the specified role of industry in carrying out such a project.

Conference on Nucleonics in Medicine to Be Held

Two full-day sessions, the first devoted to electronics in medicine, and the second to nucleonics in medicine, will constitute the Fifth Annual Conference on Electronic Instrumentation and Nucleonics in Medicine. The conference, sponsored by the American Institute of Electrical Engineers, will be held in New York City on Nov. 24 and 25, at the Hotel New Yorker.

The chairman for the morning session on nucleonics is G. Failla, department of radiology, Columbia University. The titles and authors of the papers to be given in that session are: *Isotope Measurement Techniques in Medicine*, J. W. Hitch, advisory field service branch, Atomic Energy Commission, Oak Ridge; *How to Get the Most Out of Your Scintillation Counter*, W. W. Schultz, General Electric Co., Schenectady; *Tissue Equivalent Ionization Chambers*, H. H. Rossi, Columbia University; *Light Collection and Transmission in Photoelectric Detection Devices*, Z. Collins and E. Webb, Westinghouse Electric Corp., Baltimore; and *Medical Applications of Ohmart Cells*, E. L. Saenger and J. T. Ling, radioisotope laboratory, College of Medicine, University of Cincinnati.

The chairman for the afternoon session on nucleonics is R. Chamber-

lain, department of radiology, University of Pennsylvania School of Medicine. The titles and authors of the papers to be given at that session are: *Some Biological Effects of High-Energy Radiation*, H. M. Rozendaal and W. D. Bellamy, General Electric Co., Schenectady; *Effects of Airborne Radiation on the Surrounding Environment*, J. J. Fitzgerald, Knolls Atomic Power Laboratory, Schenectady; *Techniques for Localized Irradiation and Dosimetry of the Animal Body with High-Energy Deuterons*, C. A. Tobias and H. O. Anger, Donner Laboratory, University of Californin, Berkeley; and *Technical Aspects of the High-Energy Electron Beam Used in Therapy*, J. S. Laughlin, J. Obadia, R. A. Harvey, and L. L. Haas, department of radiology, College of Medicine, University of Illinois, Chicago.

AEC Selects Goodyear to Operate New U-235 Plant

The Goodyear Tire and Rubber Co., Akron, Ohio, has been selected by the Atomic Energy Commission to operate the gaseous-diffusion plant to be built in Pike County, Ohio (NU, Sept. '52, p. 67).

As operating contractor, Goodyear will employ a permanent force of about 4,000 persons.

Radiological Health Is Subject of Series of Courses

As a part of the radiological health training program sponsored by the U. S. Public Health Service, a series of short courses is being given at the Environmental Health Center, Cincinnati, Ohio.

The training is designed for professional people who are concerned with radiological health problems in their respective fields. Candidates should have a degree in medicine, engineering, or the physical or biological sciences.

The program is divided in basic, intermediate, and advanced courses.

The basic course provides training in the theory of radiation and radiation detecting instruments, calibration and use of these instruments, detrimental effects of radiation, means of shielding and protection against radiation, and permissible exposure levels. Basic courses are scheduled to start Oct. 6, 1952, Jan. 19, and April 20, 1953.

The intermediate course covers area

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and personnel monitoring, radioassay of water, food, sewage, and biological samples, and operation, maintenance, and repair of radiation detection instruments. Intermediate courses are scheduled to start Oct. 20, 1952, Feb. 2, and May 4, 1953.

The advanced course is primarily for personnel who are concerned with occupational radiological health problems. The course covers present and probable future applications of radiation in industry and certain institutions. Radiation phenomena, associated health hazards, recommended protection principles, and applied instrumentation are also discussed. This course is scheduled to begin Feb. 16, 1953.

Further details may be obtained from the Chief, Radiological Health Training Section, Public Health Service, Environmental Health Center, 1014 Broadway, Cincinnati 2, Ohio.

Raw Materials

• **Utah.** The Bullion Monarch Mining Co. is going to search for uranium ore in Utah, under a contract with the Defense Minerals Exploration Administration.

A uranium ore buying and sampling plant with a 200-ton/day capacity will be built in Elgin, Utah.

• **Wyoming.** The Homestake Mining Co. has discovered two small deposits of high-grade uranium ore in the northeastern part of the state. The ore is carnotite in sandstone formations.

• **Uranium recovery unit.** The first commercial unit for recovery of uranium from phosphate rock has begun operation at the phosphate-chemicals plant of the Blockson Chemical Co. near Joliet, Ill. The company has a contract to supply the recovered uranium to the Atomic Energy Commission.

Uranium is a minor component of the extensive phosphate deposits of Florida and several Western states. A ton of phosphate rock contains about 0.2–0.4 lb of uranium. Over 9-million tons of rock are mined annually. AEC-sponsored studies have established the feasibility of recovering this uranium as part of the manufacturing process for sodium phosphate chemicals and concentrated commercial fertilizers.

Other full-scale units for recovering uranium from phosphate rock are being built by International Minerals and Chemical Corp. and the Virginia-

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IN BRIEF

• **Rocky Flats.** The major part of construction of the Rocky Flats, Colo., project will be completed early in 1953, well ahead of schedule, according to Admiral G. C. Hoover, project field manager. The plant will work with radioactive materials, but there will be no weapons manufacture or production, it is said.

• **Reactor in Sweden.** The Swedish government has allocated about 3-million kronor for atomic research. Most of the allocation will be used for purchasing heavy water from Norway. A 100-kw, heavy-water moderated reactor is under construction near Stockholm.

NUCLEAR NEWSMAKERS

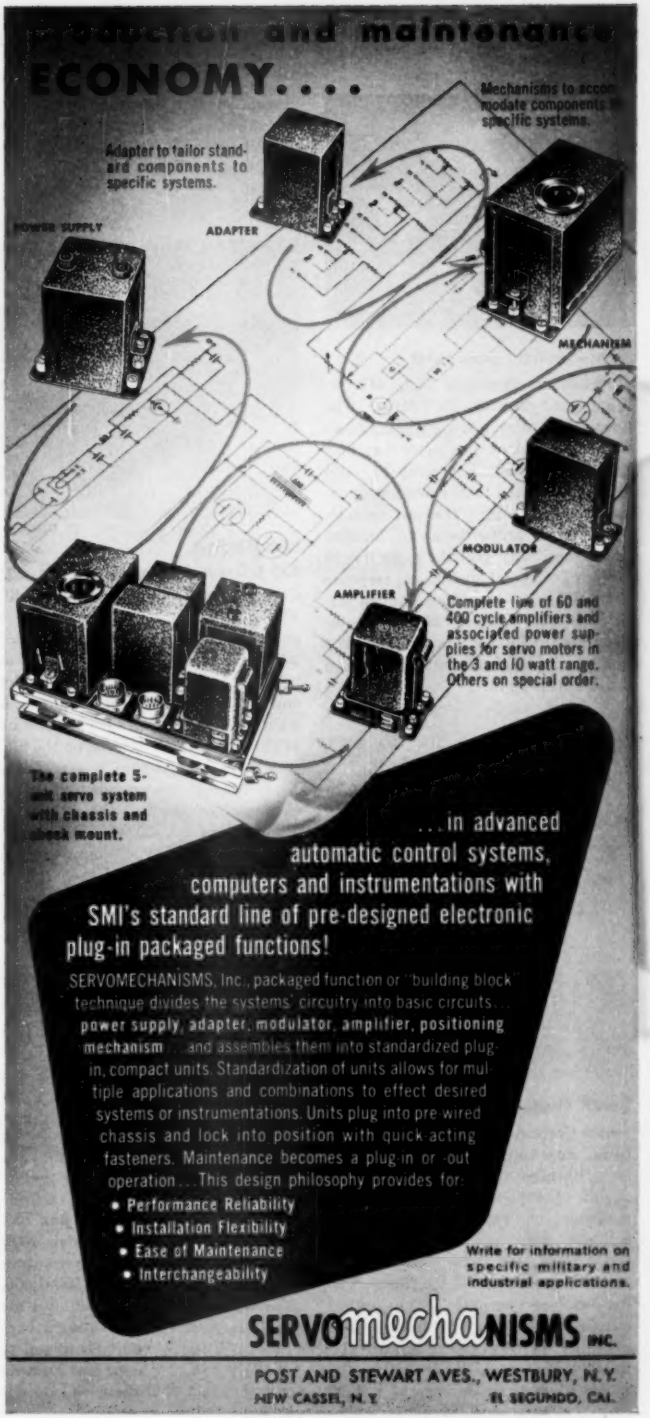
Granvil C. Kyker, head of the department of biochemistry and nutrition, School of Medicine, University of Puerto Rico, has been appointed principal scientist of the medical division of the Oak Ridge Institute of Nuclear Studies. He succeeds **H. D. Bruner** who has become chairman of the Department of Physiology, Emory University Medical School.

Urner Liddel has joined the staff of Bendix Aviation Corp. in Detroit as a director of product development. Dr. Liddel has been chief, physics branch, AEC, and, prior to that, director, physical sciences division, ONR.

Arthur W. Waltner, associate professor of physics at North Carolina State College is spending this year at the University of Stockholm working on the design and construction of the heavy-water, natural-uranium reactor being built there. In exchange, **Pelle Isberg** of the Swedish group is spending the year at North Carolina State assisting in construction and calibration of the 10-kw enriched reactor.

Jesse D. Perkinson, Jr., senior scientist, medical division of ORINS, has joined the staff at the University of Tennessee Medical Units as associate professor in the Department of Biochemistry.

Gonzalo Segura has been appointed chief radiochemist and will direct the radiology department of Foster D. Snell, Inc.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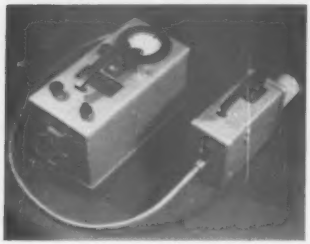
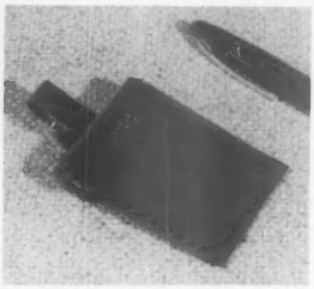


Fast-Neutron Dosimeter

Radioactive Products, Inc., 443 W. Congress, Detroit 26, Mich. The model E-1, proton-recoil-proportional-type fast-neutron dosimeter has four ranges: 0–5, 50, 500 mrep/hr, and 0–200 integrated total count (approximately 0.5 mrep). The instrument's response is proportional to human-tissue sensitivity over the range 0.2–10 Mev. It has 2:1 directional response and 10:1 pulse-amplitude gamma discrimination. Battery life is greater than 200 hr for continuous use. The combined weight of battery box and hand unit is 10 lb.

Small Photomultiplier

Radio Corporation of America, Tube Dept., Harrison, N. J. The RCA-6199 is a 10-stage tube with an over-all length of 4 9/16 in. and a maximum diameter of 1 9/16 in. Its 1¼-in. cathode, with an S-4 response, is located under a 1-in. diameter flat glass face on the end of the tube. The flat surface permits good optical coupling between a scintillation phosphor and the tube. Over-all cathode-to-anode voltage is 1,250 volts maximum; average anode current is 0.75 ma. The tube provides current amplification of 6×10^5 with an average equivalent anode-dark-current input of 8×10^{-10} lumen with a 1,000-volt test supply voltage.



Film Badge

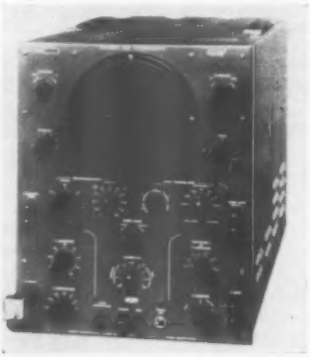
R-C Scientific Instrument Co., Inc., 335 Culver Blvd., Playa Del Rey, Calif. The Dosifilm film badge employs a polyvinyl-chloride envelope, clamped into a nickel-plated metal closure to protect the film packs against water and light. An identification number is stamped sufficiently deep on the closure so that it is directly printed on the film pack when the closure is exposed to X-ray. Loops are provided inside the envelope for holding the lead-silver filter (NU, July, '50, p. 26) in place. Dimensions are 1½ × 2 in., excluding the clip; weight of the fully loaded badge is ½ oz.

D-C Power Supplies

John Fluke Engineering Co., Box 755, Springdale, Conn. The models 400B and 400C d-c power supplies have output voltages of 1,000–5,000 and 500–1,500, respectively. Both are rated at 1 ma and hold their output voltages constant to ±0.01%, short term, and to ±0.1% per day. Noise and hum do not exceed 0.01% of the lowest output voltage.

Low-Cost Scaler

Tracerlab, Inc., 130 High St., Boston 10, Mass. Scaling factors of 32, 64, 128, and 256 can be selected to drive the register of the SC19 scaler. A preset-count switch will stop the register at 10, 100, or 1,000 for predetermined counts. The maximum counting rate of the register is 1,000 cpm; coincidence loss up to 60,000 cpm is less than 1%. A separate clock or preset timer can be connected to the scaler. The unit includes a high-voltage power supply for G-M tubes; high voltage varies 0.1% for 1% change in line voltage.



Oscillograph

Instrument Division, Allen B. Du Mont Laboratories, Inc., 1500 Main Ave., Clifton, N. J. The type 303A wide-band, high-gain, cathode-ray oscillograph is designed for study of pulses and other high-speed phenomena. The unit is equipped for quantitative measurement of time and amplitude. Nominal bandwidth is 10 Mc with transient response of 0.033 μsec. Gradual fall-off of Y-axis frequency response allows display of signals of 20 Mc. Sweep durations are variable

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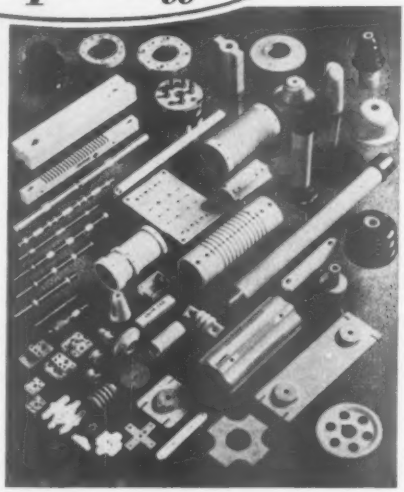

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from 0.1 sec to 2 μsec. Sweeps may be expanded, with full positioning, up to 6 times full-screen diameter. Square-wave standards of 0.1, 1.0, 10, and 100 volts, with an accuracy of ±5%, provide amplitude calibration; sinusoidal timing markers of 0.1, 1, 10, and 100 μsec have an accuracy of ±3%. The deflection factor of the vertical amplifier is 0.1 volt/in. peak to peak with an undistorted vertical deflection of 1.5 in. for unidirectional signals and 3 in. for symmetrical signals. The calibrated scale has variable illumination.

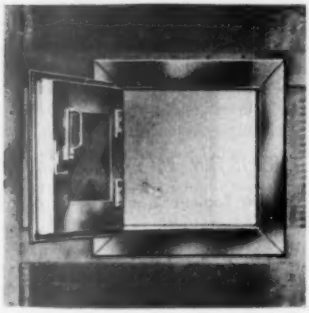
Electronic Wattmeter

Keithley Instruments, 3868 Carnegie Ave., Cleveland 15, Ohio. The model 109 electronic wattmeter employs an amplifier driving the potential coil of a dynamometer wattmeter which provides an input impedance of 500,000 ohms and eliminates errors due to potential-coil current. A response of 20–3,000 cycles/sec permits measurement at all power frequencies and their harmonics, of nonlinear circuits with harmonics, and of low audio frequencies. Watt ranges are 300, 100, 30, 10, 3, and 1 with full-scale accuracy within 2%. Maximum permissible current is 1.0 ampere; external shunts are available for measuring 3.0- and 10-ampere circuits.

Plastic Piping

American Agile Corp., Plastics Division, P. O. Box 168, Bedford, Ohio. These ducts and pipe fittings, made of polyethylene, unplasticized and rigid polyvinyl chloride, or rigid rubber-resin, are resistant to corrosive liquids and gases up to 175° F. They are light-weight and can be incorporated into existing pipe lines. All the ducts are available up to 6 in. in diameter,

and the polyethylene up to 22 in. Pipe fittings, such as tees, crosses, 45 and 90-deg elbows, are of fully-welded construction.

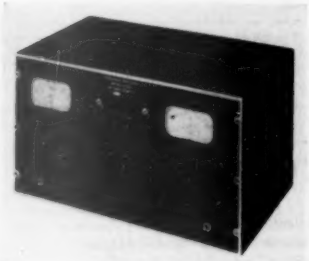


Access Door

Ace Engineering and Machine Co., 3644 N. Lawrence St., Philadelphia 40, Pa. The access door, for entrance to enclosures shielded against radiofrequencies, incorporates double-shielded construction and serrated phosphor bronze contact strip; standard sizes are 16 × 16 in. and 24 × 24 in., with other sizes available on special order. They may be added to existing installations.

Labeled Biochemicals

Charles E. Frosst and Co., P. O. Box 247, Montreal, Canada. The following radioactively labeled compounds are available: estrone 16-C^{14} (2.7 mc/mg), progesterone 21-C^{14} (2.25 mc/mg), desoxycorticosterone acetate 21-C^{14} (2.25 mc/mg).



Regulated Power Supply

Oregon Electronic Mfg. Co., 206 S. W. Washington St., Portland 4, Oregon. The model D6 heavy-duty regulated power supply provides two outputs continuously variable from 0–600 volts at a maximum current of 200 ma each. Regulation is better

NMC

from tritium to the transuranics . . .

less than 0.5% error

NMC has achieved a new and amazing precision and dependability with the PC-3 Proportional Counter employing the Candid Decade Scale of 1,000, a counting system of highest accuracy. The entire range of elements from Tritium to the Transuranics may now be recorded with less than 0.5% instrumental error. The scaling factor of 1,000 comprises three Candid Decade plug-in strips. Resolution time is 5 microseconds over all, scaler resolution, 2 microseconds, and maximum counting rate, 1,000,000 CPM.

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The PC-3 detects alpha and beta activity simultaneously (in the presence of gammas) while discriminating one from the other. The PC-3 is ideal for soft beta work. Wide applications include the fields of: radiochemistry, radiobiology, health physics, water and sewage assay, and nuclear physics.

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Geometry:	2π for alpha and beta.
Resolution loss:	1% per 100,000 c/m.
Plateaus:	Alpha, 1000 v. - 1400 v.; beta 1700 v. - 2000 v. slope 0.5% for the best 100 volts.
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CORONA NEW YORK

than 0.5% with ripple of less than 10 mv, peak to peak. The two outputs can be combined to give a maximum output of 400 ma. Other outputs are 6.3 volts, a-c, at 10 amperes, center-tapped, 0 to −150 volts, d-c, at 5 ma, variable and stabilized. Voltmeter and milliammeter may be switched to monitor any of the outputs.

Air Filter

Mine Safety Appliances Co., Braddock St., Pittsburgh 8, Pa. Each Ultra-Aire Space Filter is reportedly tested and certified to be at least 99.95% efficient against particles as small as 0.3 μ in diameter. The filter is available in sizes of 50, 500, and 1,000 ft³/min. Its effective service life, when protected by a roughing filter, is claimed to be about a year and a half. The unit can be incorporated in air conditioning systems, either in the intake or the exhaust.

Labeled Biochemicals

Schwarz Laboratories, Inc., 202 E. 44th St., New York 17, N. Y. Biochemicals are available labeled with S^{35}, C^{14}, and P^{32}. Some of the compounds available from stock or on special order are: glutathione, ribonucleic acid and derivatives, adenine nucleotides, purine bases, amino acids, sugar phosphates, and sugars. Compounds labeled with both C^{14} and P^{32} can be prepared on special orders.

Autoradiographic Stripping Film

Eastman Kodak Co., 343 State St., Rochester 4, N. Y. When stripped from the cellulosic-ester temporary support, this permeable-base stripping film has a permanent backing of 5-micron-thick gelatin. Emulsion thickness is approximately 5 microns; sensitivity for alpha and beta particles is between nuclear-track-emulsion-types NTB and NTA. The impermeable layer between the emulsion and the radioactive section avoids any artifacts by chemical action or abrasion, and prevents direct contact between the photographic developer and the section.

INDUSTRY NOTES

▶**Data Engineers,** 4608 Ravenswood Ave., Chicago 40, Ill., is a new company formed for the designing and manufacturing of data-recording equipment. A three-story building houses an engineering department, electronic laboratory, and model shop.

▶**Mathematical Computing Service** 105 Court St., Brooklyn 2, N. Y., has been organized to serve as consultants in applied mathematics to industries and universities in the fields of engineering and nuclear physics.

LITERATURE AVAILABLE

Containers. Bulletin describes containers and flasks for liquefied gases. *Superior Air Products Co., 132 Malvern St. Newark, N. J.*

Transformers. Catalog gives data on transformers and reactances. *Southwestern Industrial Electronics Co., 2831 Post Oak Road, Houston 19, Texas.*

Instruments and radiochemicals. Catalog L lists equipment for radioisotope applications. *Nuclear Instrument and Chemical Corp., 229 W. Erie St., Chicago 10, Ill.*

Tachometers. Data sheet T4 describes hand, portable, and fixed

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3116 Michigan Drive Louisville 5, Kentucky

installation tachometers. *Metron Instrument Co., 432 Lincoln St., Denver 9, Colo.*

Liquid-level controls. Illustrated catalog gives data on floatless liquid-level and industrial controls. *B/W Controller Corp., 2200 E. Maple Rd., Birmingham, Mich.*

Glassware. Brochure tells how to get specially designed scientific glassware. *Wakefield Industries Inc., 5108 W. Grove St., Skokie, Ill.*

Flowmeter. Catalog 40 describes high-pressure high-temperature flowmeters. *Fischer and Porter Co., Hatboro, Pa.*

Laboratory equipment. Illustrated catalog describes stainless metal laboratory equipment. *Kewaunee Mfg. Co., 5008 S. Center St., Adrian, Mich.*

Radioactive solutions. Brochure discusses effects of NaI^{131} solutions. *Abbott Laboratories, North Chicago, Ill.*

Air cleaner. Catalog 252 shows uses of cleaner for compressed gas and air lines. *Logan Engineering Co., 4901 W. Lawrence Ave., Chicago 30, Ill.*

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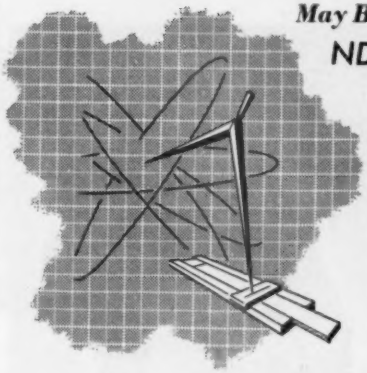

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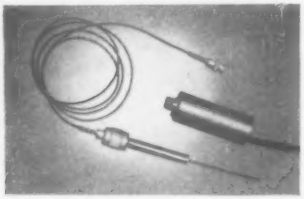

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Permit the use of Robinson-Selverstone probing counter tubes with standard commercially-available scalers. Complete information on interconnection is included.

Detailed technical data will be sent immediately on request.

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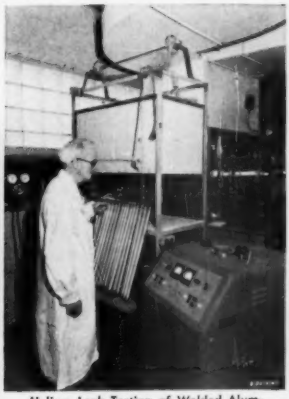
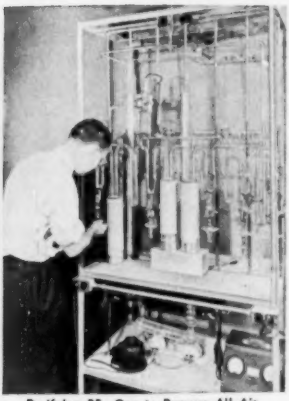
R C L

Welding End Plugs in the All Aluminum BF_3 Counter.



Helium Leak Testing of Welded Aluminum Shells before Filling with Enriched BF_3 Gas.



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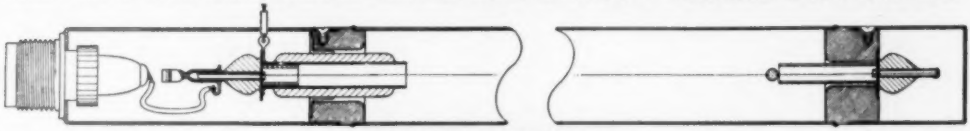

RCL MARK 2 MODEL 3

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		12 cm. 1200 v.	3% per 100 v.	139.50

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- Acetonitrile-2-C^{14}
- Bromoacetic acid-1-C^{14}
- Bromoacetic acid-2-C^{14}
- Sodium cyanoacetate-2-C^{14}
- Methyl cyanoacetate-2-C^{14}
- Diethyl malonate-2-C^{14}
- Diethyl malonate-1, 3-C^{14}
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- 2.4 D-1-C^{14}
- 3-p-methoxyphenyl-n-butyric acid-1-C^{14}
- Glycine-1-C^{14}
- Glycine-2-C^{14}
- Hippuric acid-1-C^{14}
- Hippuric acid-2-C^{14}
- DL-3-Phenylalanine-2-C^{14}
- Naphthalene-1-C^{14}
- 1-Naphthol-8-C^{14}
- 2-Naphthol-8-C^{14}
- 2-Naphthylamine-8-C^{14}
- Acetylene
- Urea
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- Acetone-2-C^{14}
- Ethanol-1-C^{14}
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